


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
課題番号 Project No. 2014A0291 実験課題名 Title of experiment Observation of the inside of a lithium-ion battery by muonic x-ray 実験責任者名 Name of principal investigator Izumi Umegaki 所属 Affiliation Toyota Central Research & Development Labs., Inc.	装置責任者 Name of responsible person Prof. Yasuhiro Miyake 装置名 Name of Instrument/(BL No.) D2 実施日 Date of Experiment 2014/5/6-2014/5/9

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.
Components of lithium-ion battery; Li(Ni <sub>0.85</sub> Co <sub>0.15</sub> )O <sub>2</sub> , as a cathode material Graphite, as an anode material Al sheets Cu sheets Polymer separator

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.
<p>This experiment is an ambitious challenge to apply an elemental analysis technique with muonic x-ray to a non-destructive compositional analysis for light elements, such as, Li, C, F, and O in multi-layers samples like commercial batteries. We have initiated to measure a depth profile of several elements in the cathode and anode sheets of a Li-ion battery with this technique.</p> <p><b>【Method】</b></p> <p>Negative mions (<math>\mu^-</math>) are implanted into samples and the implanted <math>\mu^-</math> is trapped by a nearby element, resulting in the formation of a muonic shell structure like an electronic shell structure. Then, the trapped <math>\mu^-</math> transits from a higher energy level to a lower one with emitted x-ray, whose energy corresponds to the difference in the energy between the levels. Such x-ray is usually called as a muonic x-ray. The transition occurs several times until the <math>\mu^-</math> reaches down to the lowest energy level, i.e. the <math>\mu</math>K-shell. Note that the muonic x-ray is easily detected by a Ge semiconductor detector, because the energy of the muonic x-ray is about 200 times larger than that of the x-ray fluorescence.</p>

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

In order to suppress the decrease in the intensity of the muonic x-ray, the sample was set in the Al chamber, which is evacuated with a turbo-molecular pump during the experiment. Since the Al chamber equips six windows, we used six Ge detectors. Each Ge detector was arranged close to each window to cover a large solid angle. The range of the  $\mu^-$  in the sample was adjusted by changing the implantation momentum ( $p_\mu$ ) of  $\mu^-$ .

### 【Experimental results】

At first, we prepared a LiF crystal, which is sandwiched by two Al foils. As seen in Fig.1, we have successfully observed a  $\mu\text{Li-K}\alpha$  signal at 18.70 keV, which has been never detected by an x-ray fluorescent analysis. This is, to our knowledge, the first report to detect the  $\mu\text{Li-K}\alpha$  signal.

Then, we measured the multi-layer sample consisting of an anode sheet, separator, a cathode sheet, and Al foils. Figure 2 shows an energy profile of the muonic x-ray mainly from the cathode. Since the cathode contains 5wt% carbon as a conductive additive, the profile includes the signals not only from Li and Al but also C. Such C is found to induce difficulty to detect the Li signal, because the  $\mu\text{C-L}\beta$  signal also locates at 18.83 keV, which is higher only by 130 meV than the energy of the  $\mu\text{Li-K}\alpha$ . The energy resolution of the present Ge semiconductor detector is 400 meV and too large to separate into two signals. In order to obtain information on the distribution of Li in the cathode and other components, we plan to use the detector with a higher energy resolution, such as a SDD or CdTe detector.

It is also found that the depth distribution of the implanted  $\mu^-$  becomes broader with increasing  $p_\mu$ . When we use  $p_\mu=15.7(21.2)\text{MeV}/c$ ,  $\mu^-$  stops the region from the surface with  $120\pm 20(300\pm 60)\mu\text{m}$  depth. Furthermore, since the deviation of the selected  $p_\mu$  is not zero but  $\pm 5\%$ , such distribution of  $p_\mu$  further extends the depth distribution of  $\mu^-$ . In order to analyze the composition as a function of depth, it is highly desirable to obtain “monochromatic negative muon beam”.

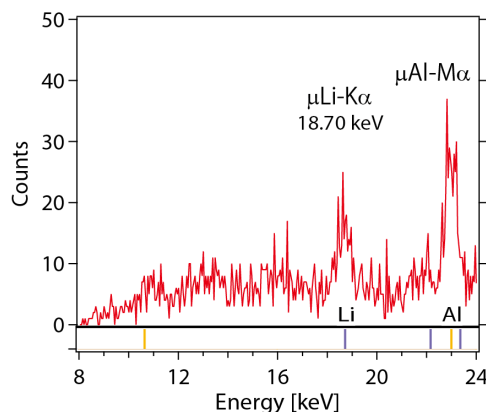


Figure 1 : Energy profile obtained for LiF crystal.

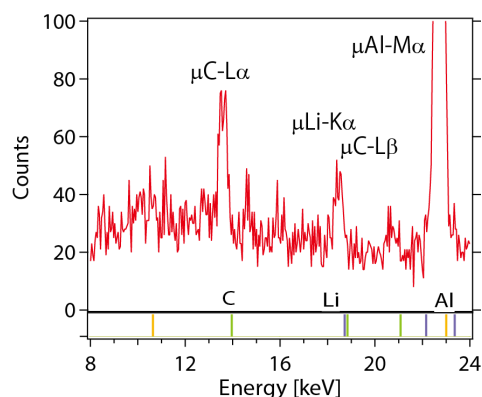


Figure 2 : Energy profile for the cathode, which consists of cathode materials and Al current collector.