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。 ）

MLF Experimental Report	提出日 Date of Report 2016.9.14
課題番号 Project No. 2015A0106 実験課題名 Title of experiment Observation of Li in 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 Laboratories, Inc	装置責任者 Name of responsible person 三宅康博 教授 装置名 Name of Instrument/(BL No.) D2 実施日 Date of Experiment 2016.5.22-2016.5.24

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.

Cathode materials of a Li-ion battery, i.e. $Li_x(Ni_{0.8}Co_{0.2})O_{2-y}$

(x, y) = (0, 0.93) (0.33, 0.58) (0.67, 0.30) (1, 0)

20 g each

These powder samples were prepared by mixing three commercially available powders, namely, $Li(Ni_{0.8}Co_{0.2})O_2$, NCo_3O_4, and NiO. The molar ratio between Ni and Co was kept at 0.8 : 0.2, while the amount of Li was changed from 0 to 1. That is, Li : Ni :Co = x : 0.8 : 0.2 with x = 0, 0.33, 0.67 and 1.

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.

In order to observe the inside of Li-ion battery non-destructively under working condition, we need to develop a compositional analysis technique with muonic x-ray, since it is a powerful tool for detecting Li. In our first attempt, we have observed a Li-Kα signal from a Li-ion battery cathode sheet. We also found that the intensity of the signal from carbon (C), which is included in additives in the cathode sheet and a polymer separator, is comparable to that from Li. Unfortunately, since the energy of the C-Kβ signal is higher than that of Li-Kα by only 130 eV, it is difficult to distinguish the latter signal from the former one with a conventional Ge semiconductor detector. This is because its energy resolution (ΔE) is 300 eV.

Then, we have prepared powder cathode materials without carbon, but with a different Li content (x) by mixing the following commercially available powders, $Li(Ni_{0.8}Co_{0.2})O_2$, NiO, and Co_3O_4 in order to obtain the Li signal only from the cathode materials. The samples were packed in an aluminum sample cell, which has a window with a size of 100 mm × 100 mm (Fig. 1). The negative muons were implanted into the cell at an angle of 45^o from the normal line. We measured muonic x-ray profiles for the four samples to obtain the evolution of the intensity of the Li-Kα signal as a function of the Li content (x).

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

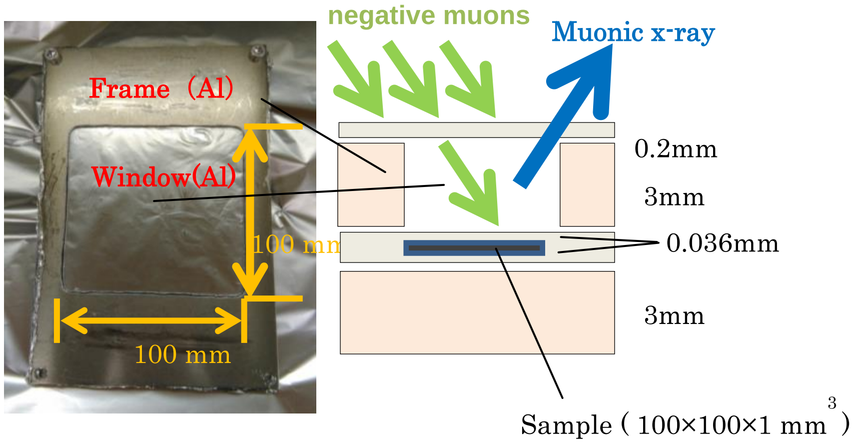

Fig. 1 (left) A picture of the front view of the sample cell. (right) A cross sectional view of the sample cell.

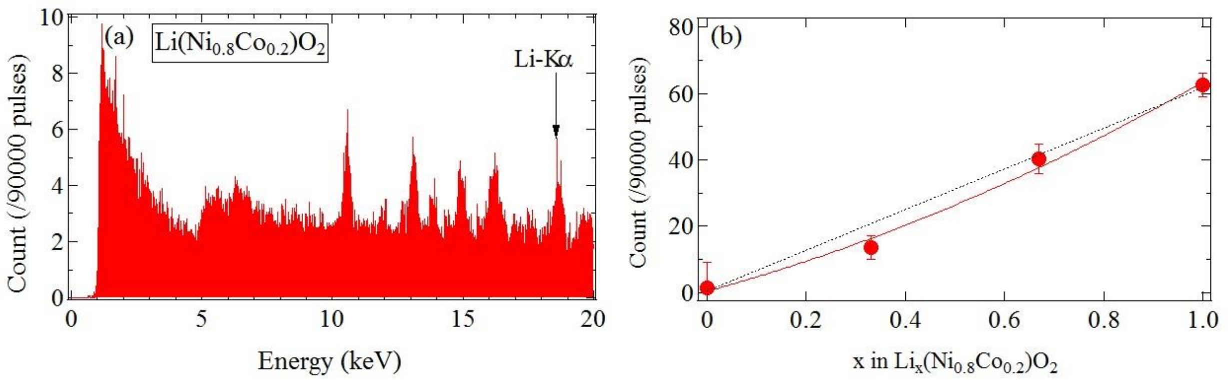

Fig. 2 (a) The muonic x-ray profile for the x=1 sample in the energy range below 20 keV. The energy resolution of the Ge detector (ΔE) is 200 eV. (b) The relationship between the Li content (x) and the intensity of Li-Kα signal.

Figure 2(a) shows the muonic x-ray profile for the x=1 sample. The intensity is normalized by 90000 pulses, which is equivalent to the pulse number in one hour of J-PARC. The Li-Kα signal is observed at 18.7 keV for the x=0.33, 0.67, and 1 samples. The intensity of the Li-Kα signal is found to increase in proportion to x [Fig. 2(b)]. This demonstrates that the Li content in the cathode materials is evaluated by this technique. On the other hand, it is required to distinguish the C-Lβ signal from the Li-Kα signal, since a Li-ion battery contains a polymer separator and cathode and anode sheets contain carbon as additives. We expect that the Li-Kα signal is separated from the C-Lβ signal using a Ge semiconductor detector with higher energy resolution (e.g. ΔE=200 eV). However, for more precise estimation of the Li content in the cathode materials, we definitely need to further develop this technique.