MLF Experimental Report	提出日 Date of Report
課題番号 Project No. 2012A0212 実験課題名 Title of experiment Structural analysis of nano-sized transitional-metal oxide as negative electrode materials for Li-ion batteries 実験責任者名 Name of principal investigator Yohei ONODERA 所属 Affiliation Kyoto University	装置責任者 Name of responsible person Toshiya Otomo 装置名 Name of Instrument/（BL No.） NOVA (BL21) 実施日 Date of Experiment 2013/3/22-24

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.

Nano-sized cobalt oxide, CoO

Mechanical alloyed LixCoO (x=0.5, 1.0, 1.5, 2.0, 2.5)

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 this proposal, we carried out neutron total scattering experiments in order to study the local structure of nano-sized Li-CoO conversion negative electrode materials, which is known as promising high-capacity and high-reversibility negative electrode materials for Li-ion batteries.

The Li_xCoO samples were synthesized by the mechanical alloying (MA) method from Li metal foil and nano-sized cobalt oxide powder. The starting materials were put into a SUS pot with 20 SUS balls under high purity argon gas atmosphere, and then alloyed mechanically at 100 rpm for 10 h using a planetary ball mill apparatus (Fritsch, P-5).

The neutron total scattering experiments were carried out with the High Intensity Total Diffractometer (NOVA), installed at BL21 of MLF in J-PARC. The powder samples by MA Li_xCoO (x=0.5, 1.0, 1.5, 2.0, 2.5), and CoO nano-sized powder were put into cylindrical vanadium-nickel cells (6 mm in diameter) under argon gas atmosphere. The neutron total scattering data of the samples were measured for 6 h at room temperature with 300 kW of MLF beam power. Structure factors, $S(Q)$, were obtained from the scattering intensity after correction of the background, attenuation, and multiple scattering and normalization with scattering from a vanadium rod.

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

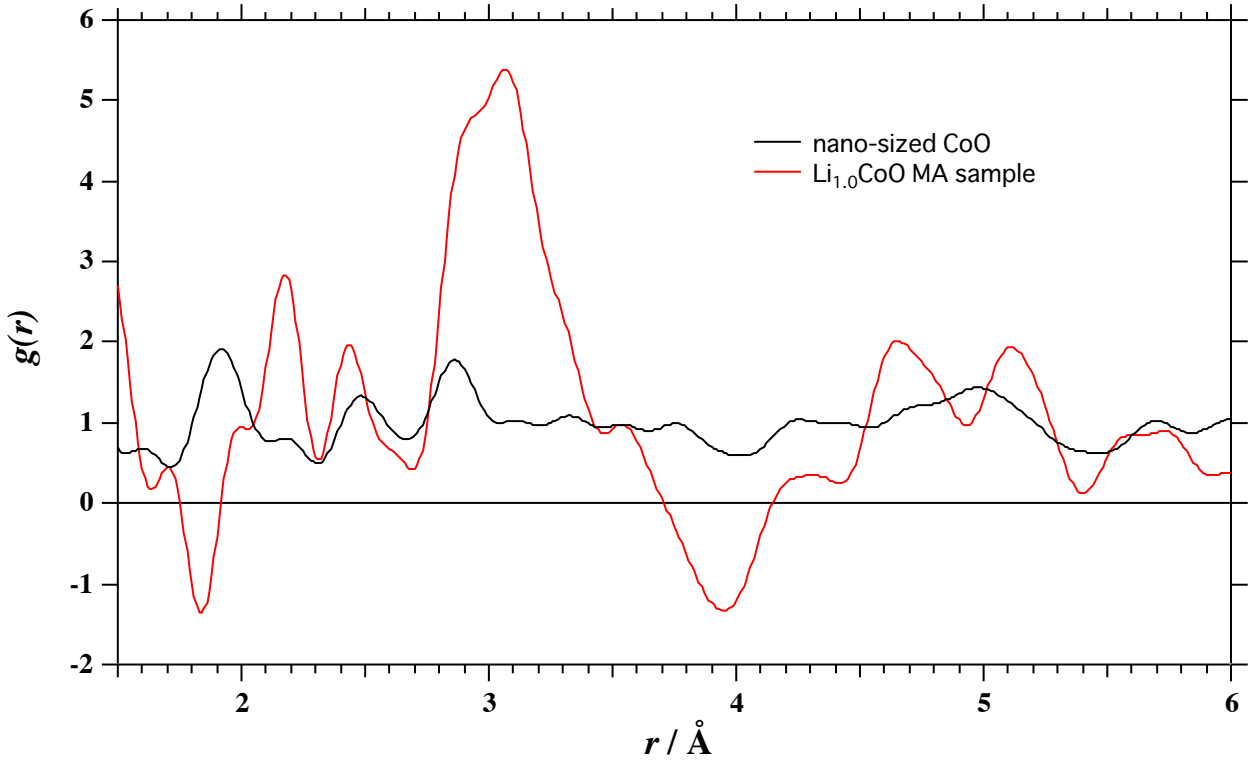

Fig.1 Pair distribution functions, $g(r)$ for Li_xCoO (x=0, 1.0) samples.

Figure 1 shows pair distribution functions, $g(r)$, obtained by the Fourier transformation of $S(Q)$s for Li_xCoO (x=0, 1.0) samples with Q range from 0.4 to 35 $Å^{-1}$. A peak was observed around at 1.9 Å in $g(r)$ for the nano-sized CoO sample and it was attributed to Co-O nearest-neighbor correlation. Co-O correlation peak was vanished and a characteristic negative peak was observed at around 1.8-1.9 Å in $g(r)$ for the $Li_{1.0}CoO$ sample. This negative correlation was attributed to Li-O correlation. Additionally, sharp positive peaks, correspond to Co-Co correlation in metallic Co, also were emerged at around 2.2 Å and 2.45 Å in $g(r)$ for the $Li_{1.0}CoO$ sample. These results suggest that the conversion reaction was occurred on MA treatment as follows.

$$CoO + 2Li \leftrightarrows Li_2O + Co$$

In the future, we will perform the detailed analysis for Co-O, Li-O, and Co-Co correlation by combination with high-energy X-ray diffraction data, and elucidate a relation between the structure and remarkable electrode properties on conversion reaction.