Isotope labeling method for depth profiling by neutron diffraction

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Reaction distribution in lithium ion batteries (LIBs) is a critical problem for high power application such as electric vehicles. We have tried to detect the depth reaction distribution in positive electrodes of LIB. The fabricated composite electrode has the $^{6}LiMn_2O_4$ and $^{7}LiMn_2O_4$ layers on the counter electrode side and current collector side, respectively. Thus obtained composite electrode was set in a laminated pouch-type cell as a working electrode. The cell was charged and a half of the lithium was extracted; one cell was charged at 0.05 C and the other cell was charged at 5 C. Immediately after the end of the charging process, the cell was disassembled in the glove box and the electrode was taken out, washed and then dried. Fig. 1 shows the neutron diffraction patterns of the pristine (before charging), 0.05C charged and 5C charged powder, showing that the lattice of $LiMn_2O_4$ shrinks with delithiation [1]. To obtain semi-quantitative information on the reaction distribution, we employed the Rietveld refinement [2-3] of these electrode samples. A reasonable fitting is obtained with the $^{6}Li/^{7}Li$ ratio of the pristine powder is 0.80, showing that the $^{7}LiMn_2O_4$ layer is somewhat thicker than the $^{6}Li_xMn_2O_4$ layer. A large reaction distribution was observed for the high rate experiment.

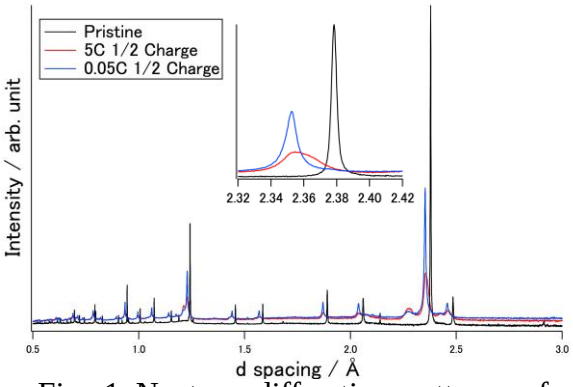

Fig. 1 Neutron diffraction patterns of pristine and charged powder.

References
[1] T. Ozuku, et. Al., J. Electrochem. Soc. 1990, 137, 769
[2] R. Oishi et. al., Nucl. Instrum. Methods A600, (2009) 94.
[3] R. Oishi et. al., J. Appl. Cryst., 45 (2012) 299.

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