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Electronic Supplementary Information for

Initial coulombic efficiency improvement of the $Li_{1.2}Mn_{0.567}Ni_{0.166}Co_{0.067}O_2$ lithium-rich material by ruthenium substitution for manganese

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Contents:

1. Experimental

1.1 Materials synthesis

The lithium-rich layered materials used in this communication were prepared by the solid state reaction technology. In a typical synthesis, firstly, required amounts of the transition metal acetates $Mn(CH_3COO)_2 \cdot 4H_2O$ (Wako), $Ni(CH_3COO)_2 \cdot 4H_2O$ (Wako), $Co(CH_3COO)_2 \cdot 4H_2O$ (Wako) and RuO_2 (Wako) were mixed thoroughly. Secondly, the stoichiometric $LiOH \cdot H_2O$ (Wako) (5% excess) were mixed with the transition metal mixture together, then calcined in the furnace at 500 ℃ for 5 h, and then the powder were pressed into pellets and calcined in the furnace at 900℃ for 15 h. Finally, these lithium-rich layered materials of $Li_{1.2}Mn_{0.567-x}Ru_xNi_{0.166}Co_{0.067}O_2$ (x = 0, 0.03, 0.05 and 0.07) were prepared, which were labeled with Ru-0, Ru-0.03, Ru-0.05 and Ru-0.07 respectively.

1.2 Characterization

The crystal structures of the pristine powders were obtained by powder X-ray diffraction (XRD) on a Bruker D8 Advance diffractometer using Cu K_α radiation. The diffraction data were recorded in the 2θ range of 10-90° with a step of 0.02° and a count time of 1s. The morphology and size of prepared powders were also observed using scanning electron microscopy (SEM, TOPCON DS-720 instrument).

1.3 Electrochemical test

The charge/discharge tests were carried out using the CR2032 coin-type cells, consisting of a cathode and lithium metal anode separated by a Celgard 2400 porous polypropylene film. The electrolyte is 1M $LiPF_6$ in ethylene carbonate/dimethyl

carbonate (EC/DEC, 1:1(v/v)). The cathode slurry were made with a weight to weight ratio of 80% of the synthesized $Li_{1.2}Mn_{0.567-x}Ru_xNi_{0.166}Co_{0.067}O_2$ (x =0, 0.03, 0.05 and 0.07) electrode materials, 15% of teflonized acetylene black (AB), and 5% of polytetrafluoro ethylene. The electrodes were produced by pressing the slurry onto a 10 micron aluminum mesh current collector and dried at 80 ℃ for 10 h in a vacuum oven. The cells were assembled in a glove box filled with dried argon gas. The cells were charged and discharged in the voltage range of 2.0-4.4 V and 2.0-4.8 V at a constant current density of 5 mA/g at 25 ℃. The electrochemical impedance spectroscopy (EIS) was performed on the Solartron 1253B Frequency Response Analyzer. The amplitude of the a.c. signal was kept at 5 mV. The frequency range of measurement was 0.6MHz-0.01Hz. Data acquisition and analysis were done respectively using the electrochemical impedance software, ZPlot and Zview.

2. Electrochemical impedance spectroscopes (EIS) of the cells with $Li_{1.2}Mn_{0.567-x}Ru_xNi_{0.166}Co_{0.067}O_2$ (x = 0.00, 0.03, 0.05 and 0.07) electrode materials.

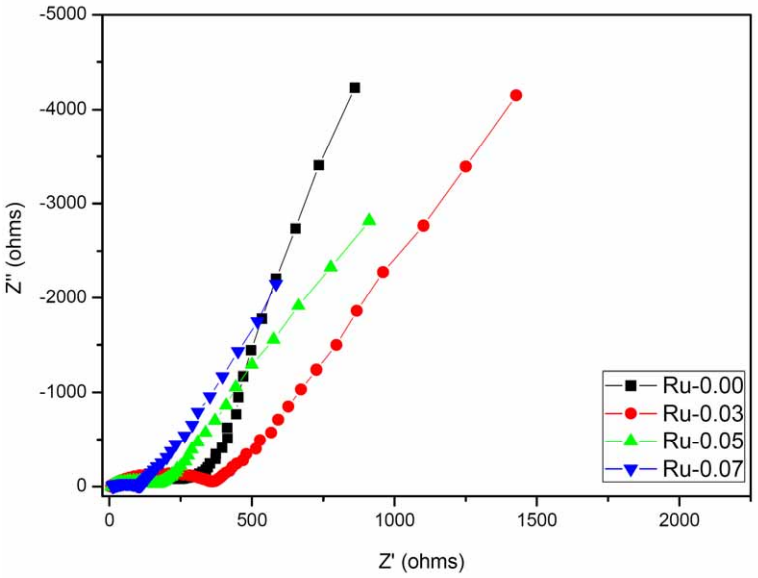

Fig. S1 Nyquist plots of the cells with $Li_{1.2}Mn_{0.567-x}Ru_xNi_{0.166}Co_{0.067}O_2$ (x = 0.00, 0.03, 0.05 and 0.07) lithium-rich layered materials prepared by solid state reaction method

3. DQ/dV curves associating with the Li_2MnO_3 component activation process.

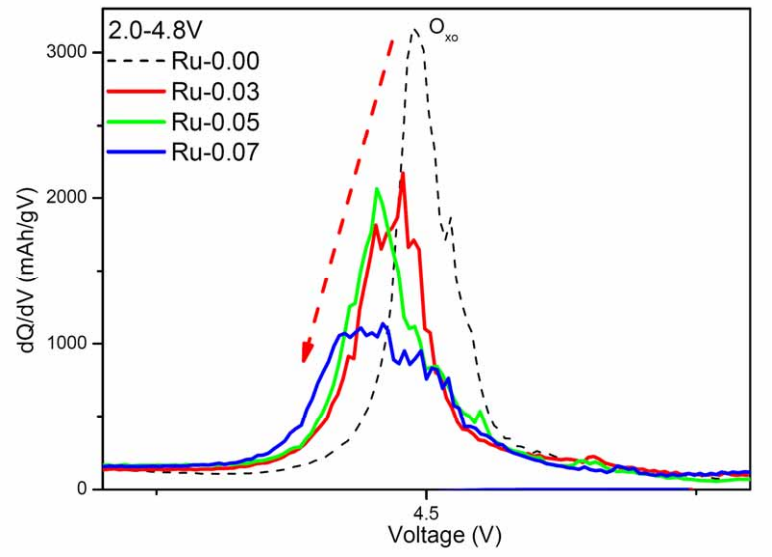

Fig. S2 Enlarged dQ/dV curves in Fig. 2(d) associating with the Li_2MnO_3 component activation process during the 1st cycle of the $Li_{1.2}Mn_{0.567-x}Ru_xNi_{0.166}Co_{0.067}O_2$ (x = 0.00, 0.03, 0.05 and 0.07) lithium-rich layered materials with 4.8V cut-off voltages.