

## Supplementary Information

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Xin Sun<sup>a</sup>, Yi Jin<sup>b</sup>, Chen-Yu Zhang<sup>a</sup>, Jian-Wu Wen<sup>a</sup>, Yu Shao<sup>a</sup>, Yong Zang<sup>a</sup> and Chun-

Hua Chen<sup>\*a</sup>

<sup>a</sup> CAS Key Laboratory of Materials for Energy Conversion, Department of Materials Science and Engineering & Collaborative Innovation Center of Suzhou Nano Science and Technology, University of Science and Technology of China, Anhui Hefei 230026, China. Fax: + 86 (0)551 3601592; Tel: + 86 (0)551 3606971; E-mail: [cchchen@ustc.edu.cn](mailto:cchchen@ustc.edu.cn)

<sup>b</sup> China Electric Power Research Institute, Department of Electric Engineering and New Materials, Beijing 100192, China

### Experimental Section

Stoichiometric amounts of NaNO<sub>3</sub>, Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O, Fe(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O, Mn(CH<sub>3</sub>COO)<sub>2</sub>·4H<sub>2</sub>O and tetrabutyl titanate were dissolved in deionized water to obtain aqueous solutions (0.2 M), into which HNO<sub>3</sub> (2 vol%) was added to avoid hydrolyzation. Then acrylic acid (AA) was added into the solutions with AA/H<sub>2</sub>O (1:2, v/v) as mixed solvents. The solutions were heated at 150°C for 10 h to complete the thermopolymerization reactions. The products were first preheated at 500°C for 10 h to get rid of poly(acrylic acid), and a subsequent heat treatment was carried out at 900°C for 10 h. All the heat treatment processes were carried out in air atmosphere.

The electrochemical properties of the layered cathode materials were evaluated in 2032 type coin cells using a Na disk as the counter electrode and 1.0 M NaClO<sub>4</sub> in PC solution as the electrolyte. The cathode laminates were prepared by mixing the active materials (84wt %), acetylene black (8wt %) and PVDF (8wt %) in N-methylpyrrolidinone into an electrode slurry and then casting the slurry on aluminium foil collector. The cells were assembled in an argon-filled glove box with above electrode laminates as working electrodes. The separator was glass fiber (Whatman Gf/D). The cells were tested on a multi-channel battery test system (NEWARE BTS-610) with galvanostatic charge and discharge in different selected voltage ranges. Cyclic voltammetry and AC impedance spectroscopy measurements on the cells were performed with a CHI 604B electrochemical workstation. The crystalline structures of the samples were characterized by using a diffractometer (Philips X'Pert Pro Super, Cu K $\alpha$  radiation). The XRD spectra were collected in a range of 2 $\theta$  values from 10° to 70° at a scanning rate of 10° min<sup>-1</sup>. The morphologies of the samples were studied by scanning electron microscopy (Sirion 2000, FEI).

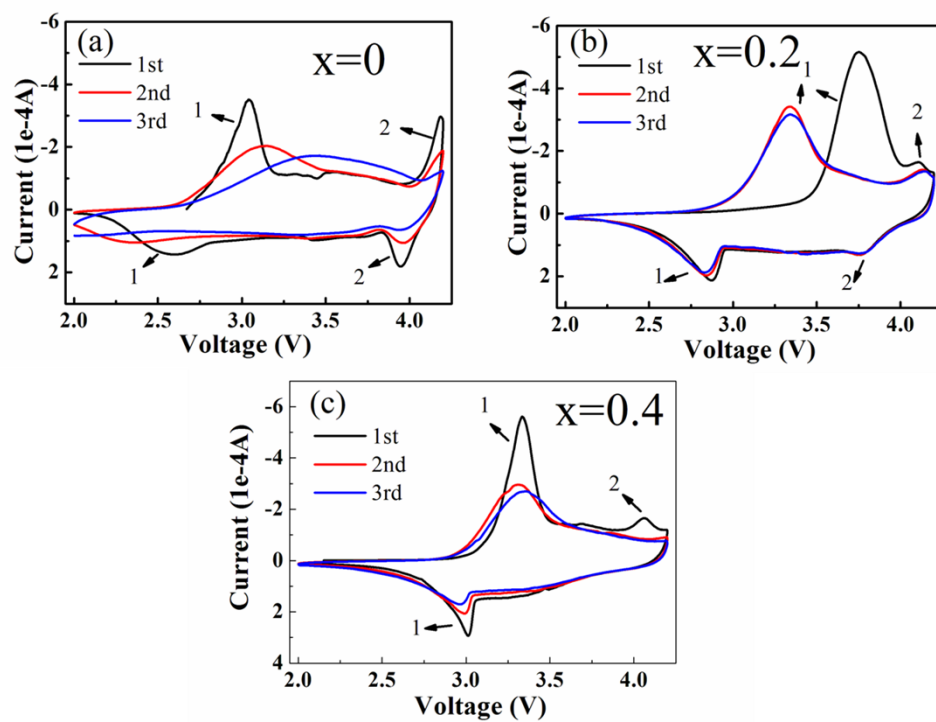


Fig. S1 CV curves of the first three cycles for cells Na/Na[ $\text{Ni}_{0.4}\text{Fe}_{0.2}\text{Mn}_{0.4-x}\text{Ti}_x$ ]O<sub>2</sub> ( $x = 0, 0.2, 0.4$ ) in the voltage range of 2 - 4.2 V and at a scan rate of 0.1 mV s<sup>-1</sup>.