## Atomic Observation on Alternating Heteroepitaxial Nanostructures in Na-ion Layered Oxide Cathodes

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Li-ion batteries (LIBs) has been used as large energy storage device over decades. Due to the increasing costs and limited Li resources, Na-ion batteries (NIBs) with similar storage mechanisms to LIBs are potentially able to be the next generation electrochemical energy storage device [1]. Layered oxide compounds with the general formula of Na<sub>x</sub>TMO<sub>2</sub>(TM = Mn, Co, Ni, etc.) can provide two dimensional (2D) diffusion path for Na intercalation/deintercalation [2,3]. However, NIBs suffer from the low capacity and cyclability, and people are devoting to solve these problems [4]. Here we report a novel layered NIBs cathode, which combine the Li<sub>2</sub>MnO<sub>3</sub> with the Na-P3 phase. This heteroepitaxial nanostructure cathode can improve the rate performance and higher initial capacity.

By the high-resolution scanning transmission electron microscopy (STEM) combined with energy X-ray dispersive spectroscopy (EDS), we can clearly distinguish the different elements. A double-aberration corrected FEI Titan G2 60-300 STEM, operated at 300 keV, was used. The screen current was 40 pA, and the convergent angle was 20.5 mrad. The EDS was acquiring using the Bruker Super-X<sup>TM</sup> technology with four detectors. To see the Li distributions, the electron energy loss spectroscopy (EELS) was recorded using a Gatan Imaging Filter (GIF) spectrometer attached to the microscope.

Figure 1.a shows the elemental mapping of Na-P3/Li<sub>2</sub>MnO<sub>3</sub> cathodes, which can be distinguished by the contrast of concentration of different elements. It can be clearly see the Na-P3 phase in elements mapping of Co, Ni and Na in brighter contrast, and the Mn element are gathering in Li<sub>2</sub>MnO<sub>3</sub>, which has lower contrast in HAADF images. To confirm the Li distributions, Figure 1.b shows the Li K-edge mapping by EELS. The lower contrast area in STEM images, which represents the Li<sub>2</sub>MnO<sub>3</sub>, shows the aggregation area of Li elements. By STEM imaging with spectroscopy technologies, the alternating heteroepitaxial structure can be easily seen and this explains to the unique electronic performance of the Na-P3/Li<sub>2</sub>MnO<sub>3</sub> cathode.

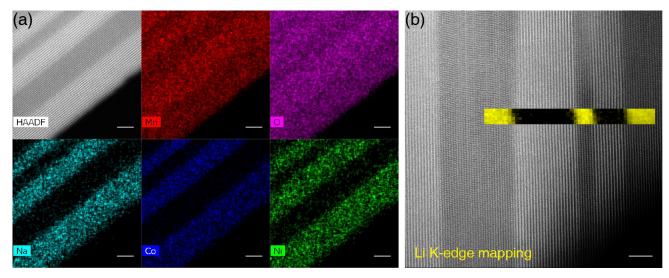
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**Figure 1.** (a) STEM-EDS mapping, (b) EELS mapping of Li K-edge. The brighter contrast areas in STEM-image corresponds to the Na, Co and Ni elements mapping, while the lower ones are connected with the Mn, O and Li elements mapping. The scale bar is 5 nm.