Mixed Layered Cathode Material with Superior Performance for Application in Na-ion Batteries

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Sodium-ion batteries (SIBs) are considered as promising candidates for cost-efficient stationary energy storage systems. Sharing the working principle and design with the already commercialized lithium-ion battery facilitate their transfer into application. However, differences in cell chemistry request the investigation of new, high performance electrode materials.

In previous work, we have focused on the development and characterization of layered Na_xMO_2 (M= transition metal) materials[1–3]. The main target was to investigate abundant, low cost and environmentally friendly elements like iron (Fe), manganese (Mn) and magnesium (Mg) to replace the expensive and toxic cobalt (Co), thus underlining the philosophy of SIBs.

In this work, we present the synthesis and characterization of novel quaternary layered cathode materials of the type $Na_xMn_yNi_zFe_{0.1}Mg_{0.1}O_2$ ($0.67\leq x\leq 1.0$; $0.5\leq y\leq 0.7$; $0.1\leq z \leq 0.3$). By tuning the synthesis conditions, pure P2- and O3-type phases as well as a mixed P-/O-type material can be obtained as proven by X-ray diffraction (XRD) and scanning electron microscopy (SEM). All materials revealed good electrochemical performance but the mixed phase material, $Na_{0.76}Mn_{0.5}Ni_{0.3}Fe_{0.1}Mg_{0.1}O_2$, revealed extraordinary performance. The high average discharge potential (3.4 V vs Na/Na^+), high capacity (128 mAh g^{-1} at 0.1C) and extraordinary capacity retention (90.2 % after 600 cycles) in a narrow potential range (2.5-4.3 V), reveal its suitability for Na-ion cells using hard-carbon as negative electrode.

We propose that the superior performance is due to the co-existence of the O-type phase (enabling high (initial) specific capacities) and P-type phase (enabling improved high rate capability as well as high-voltage stability)[4]. Most important is that the synthesis was conducted under equilibrium conditions[5] resulting in the formation of P- & O-type phases, each in its most stable elemental composition.

Further investigations are needed to clarify and explain such outstanding electrochemical performance. In any case, the unexplored class of P-/O-type mixed phases introduces various new perspectives for the development of layered cathode materials and powerful Na-ion batteries.

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