Controlled synthesis of hierarchically layered cathode materials for advanced lithium ion batteries

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Developing high-power and high-energy lithium ion batteries (LIBs) is of great technological importance for large-scale applications such as electrical vehicles (EVs), hybrid electrical vehicles (HEVs) and power-to-grid applications [1,2]. Layer-structured \( x \text{Li}_2\text{MnO}_3\)\((1-x)\text{LiMO}_2\) (M = Ni, Co, Mn, 0 \(\leq x \leq 1\)) oxides have a great potential for the use as cathode materials in such systems [3–5]. Unfortunately, some challenges remain for these materials. An obvious problem is the poor rate capability because of the low ionic and electronic conductivity of the bulk materials. Herein, layered hierarchical compounds, which are assembled from high electrochemically active \(\{010\}\) nanosheets, have been fabricated by a modified hydroxide co-precipitation route on a large scale. The as-synthesized materials with this peculiar architecture deliver an excellent electrochemical performance, especially an ultra-high rate capability. Inspiringly, the layered Li\(_{1.2}\)Ni\(_{0.2}\)Mn\(_{0.6}\)O\(_2\) cathode material exhibits high specific discharge capacity of 130 mAh g\(^{-1}\) at 10 C (1 C = 300 mA g\(^{-1}\)) between 2.0 and 4.8 V, which is superior to most lithium rich cathodes. A single layered monoclinic structure with a symmetry of \(C2/m\) was refined from neutron and synchrotron radiation powder diffraction data. Furthermore, in situ synchrotron radiation powder diffraction and in situ X-ray absorption spectroscopy were performed to investigate the (de)lithiation mechanism of the Li\(_{1.2}\)Ni\(_{0.2}\)Mn\(_{0.6}\)O\(_2\) cathode material during charge and discharge process. The outstanding electrochemical performances of hierarchically layered intercalation compounds, coupled with the scalable and universal approach, results in a high industrialization potential.

References: