

Nano-in-bulk structural cathode materials for high-performance Li ion batteries

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The electrochemical performance of cathode materials in lithium ion batteries (LIBs) strongly depends on their geometrical morphology and size. When the morphology of cathode material is altered during charging/discharging cycles, it is known that the electrochemical performance becomes severely deteriorated because the change of geometrical morphology during the cycling can continuously expose a fresh surface of cathode materials in contact with electrolyte and subsequently result in rebuilding solid electrolyte interface (SEI) layers. This geometrical variant can eventually cause an irreversible loss of capacity [1,2], thus highly necessitating effective means to control the geometrical factors like size distribution and their 3D architecture. In this study, we tried to incorporate nano-sized cathode materials with high-specific surface area into the same bulky materials by simple microwaves assistance, thereby producing a core shell structure with a bimodal size distribution. The geometrically-optimized cathode material is revealed to effectively reduce the reaction area with the electrolyte with a larger capacity and superior durability. To examine three-dimensional primary particle distribution inside a large single secondary particle, we reconstructed 3D structures of the samples using electron tomography based on FIB serial sectioning technique. The correlated atomic structures and the related chemical changes at the interfaces of SEI were uncovered via scanning transmission electron microscopy (STEM) combined with electron energy loss spectroscopy (EELS).

- [1] J. Shin et al., Morphological Evolution of Carbon Nanofibers Encapsulating SnCo Alloys and Its Effect on Growth of the Solid Electrolyte Interphase Layer, *ACS Nano* 7, 7330-7341 (2013).
- [2] Y. J. Hong et al., One-Pot Facile Synthesis of Double-Shelled SnO: Yolk-Shell-Structured Powders by Continuous Process as Anode Materials for Li-ion Batteries, *Adv. Mater.* 25, 2279-2283 (2013).