

Extending the life-span of O3-type layered oxide cathode enabled by the nanoscale aluminum oxide coating for high-energy density sodium-ion batteries

Jang-Yeon Hwang^a, Sang-Min Park^a, Yang-Kook Sun^{a,*}

^a Department of Energy Engineering, Hanyang University, Seoul, 04763, Republic of Korea

E-mail: ghkdwkd@hanyang.ac.kr

Sodium-ion batteries (SIBs) appear to be a promising alternative due to the abundance of sodium in the Earth's crust. [1] Also, the chemistry of SIBs is similar to that of well-established lithium batteries, which adds additional merit. So far, various chemical compositions have been introduced through different transition metal substitutions (Ni, Co, Mn, and Fe) such as Na[Ni_xFe_yMn_z]O₂ or Na[Ni_xCo_yMn_z]O₂ compound. [2-4] However, the O3-type layered oxides in SIBs exhibit a low reversible capacity and poor cycle retention due to the structural instability that arises from multi-phase transitions.

A surface-modified O3-type Na[Ni_{0.6}Co_{0.2}Mn_{0.2}]O₂ cathode was synthesized by Al₂O₃ nanoparticle coating using a simple dry ball-milling route. The nanoscale Al₂O₃ particles (15 nm in diameter) densely covering the spherical O3-type Na[Ni_{0.6}Co_{0.2}Mn_{0.2}]O₂ cathode particles effectively minimized parasitic reactions [5] with the electrolyte solution while assisting Na⁺ migration. The proposed Al₂O₃ coated Na[Ni_{0.6}Co_{0.2}Mn_{0.2}]O₂ cathode exhibited a high specific capacity of 151 mA h g⁻¹, as well as improved cycling stability and rate capability in a half cell. Furthermore, the Al₂O₃ coated cathode was scaled up to a pouch-type full cell using a hard carbon anode that exhibited a superior rate capability and capacity retention of 75% after 300 cycles with a high energy density of 130 W h kg⁻¹. In addition, the postmortem surface characterization of the cathodes from the long-term cycled full cells helped in identifying the exact mechanism of the surface reaction with the electrolyte and the reason for its subsequent degradation and showed that the nano-scale Al₂O₃ coating layer was effective at resolving the degradation pathways of the cathode surface from hydrogen fluoride (HF) attack.

References:

- [1] J.-Y. Hwang, S.-T. Myung, S.-K. Sun, *Chem. Soc. Rev.*, 46 (2017) 3529–3614.
- [2] J.-Y. Hwang, C. S. Yoon, I. Belharouak, Y.-K. Sun, *J. Mater. Chem. A*, 4 (2016) 17952–17959.
- [3] S.-M. Oh, S.-T. Myung, J.-Y. Hwang, B. Scrosati, K. Amine, Y.-K. Sun, *Chem. Mater.*, 26 (2014) 6165–6171
- [4] J.-Y. Hwang, S.-M. Oh, S.-T. Myung, K. Y. Chung, I. Belharouak, Y.-K. Sun, *Nat. Commun*, 6 (2014) 6856.
- [5] Y.-K. Sun, M.-J. Lee, C. S. Yoon, J. Hassoun, K. Amine, *Adv. Mater.*, 24, (2012) 1192-1196.