

Discovering the direct reason of voltage decay in Li/Mn-rich cathode material

Seungjun Myeong^a, Woongrae Cho^a, Wooyoung Jin^a, Jaeseong Hwang^a, Youngsin Yoo^a, Gyutae Nam^a, Haesung Jang^a, Jeonggu Han^a, Mingyu Kim*^b and Jaephil Cho*^a

^a *Department of Energy Engineering and School of Energy and Chemical Engineering, Ulsan National Institute of Science and Technology (UNIST), Ulsan 689-798, South Korea*

^b *Beamline Research Division, Pohang Accelerator Laboratory, Pohang, Korea 790-784*

E-mail: hjs9544@unist.ac.kr

Li/Mn-rich layered oxide materials ($\text{Li}_{1+x}\text{Ni}_y\text{Co}_z\text{Mn}_{1-x-y-z}\text{O}_2$ > 250 mAh g⁻¹) are the most promising cathode materials for high energy densities. However, they suffer from severe voltage decay upon cycling, which decreases energy densities hindering their further research and development. Here, we reveal a mechanism of voltage decay phenomenon by using conventional R-3m+C2/m($\text{Li}_{1.15}\text{Ni}_{0.17}\text{Co}_{0.17}\text{Mn}_{0.51}\text{O}_2$) compounds with capacities of ~ 250 mAh g⁻¹ and achieve voltage decay alleviation without the loss of high discharge capacities by arranging atomic composition. A combination of transmission electron microscopy and in/ex-situ X-ray absorption spectroscopy reveals that the nature of electronic/atomic structures and redox mechanisms contributes to specific TM-O/TM bonding stability, determining the rate of voltage decay in Li/Mn-rich materials. The finding advances the commercialization of Li/Mn-rich layered oxides and provides insights into structural deterioration chemistry of the cathode materials to be explored for developing high-energy densities electrodes.