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**Discovery of a Surface Protective Layer: A New Insight into Countering  
Capacity and Voltage Degradation for High-Energy Lithium-Ion Batteries**

Dong Luo<sup>a</sup>, Shaohua Fang<sup>a\*</sup>, Qinghua Tian<sup>a</sup>, Long Qu<sup>a</sup>, Li Yang<sup>a,b\*</sup>, Shin-ichi Hirano<sup>b</sup>

*a.* School of Chemistry and Chemical Engineering, Shanghai Jiao Tong University, Shanghai 200240, China. E-mail: housefang@sjtu.edu.cn (S.H. Fang) or liyangce@sjtu.edu.cn (L. Yang).

*b.* Hirano Institute for Materials Innovation, Shanghai Jiao Tong University, Shanghai 200240, China

**ABSTRACT:** Capacity and voltage degradation is a crucial factor that restricts the commercialization of Li-rich layered cathode materials. Recently, it has been demonstrated that the degradation results from the structural evolution from layered to spinel-like phase, caused by the migration of transition metal (TM) ions. However, the direction and order of TM ions migration is hard to identify. In this study, a surface protective layer (SPL) is discovered for the first time at the outer edge of  $\text{Li}_{1.14}\text{Mn}_{0.48}\text{Ni}_{0.19}\text{Co}_{0.19}\text{O}_2$  nanoplate by the research on the surface condition of nanoplates after charge-discharge cycles. More importantly, by the detailed analysis for the SPL, we further reveal that the formation of SPL is active facet dependent, TM ions migrate toward the active facets, and Ni and Co ions hop more preferentially than Mn ions during the cycling. These discoveries can help to understand the fading

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