Understanding Depth-dependent Cathode Electrolyte Interphase on the Layered Li-ion Cathodes Operated at Extreme Environment

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With the current Li-ion battery technology, the batteries can be operated between room temperature to 55 ºC, and operation beyond this suggested temperature range will lead to irreversible degradation often resulting in low cell capacity, cycle life, and sometimes catastrophic failures such as fires and explosions. However, several industrial applications require high-performance rechargeable batteries operated in aggressive environments such as military applications, sensor applications, and downhole drilling applications.[1] Fundamentally, the high-temperature operation of Li-ion batteries is highly dependent on the stability of the electrode and electrolyte interface during lithiation/delithiation electrochemical reactions (charge/discharge). However, current knowledge on the nature of cathode electrolyte interphase (CEI) formed on cathodes at extreme temperature is limited, and its stability under extreme temperature is not well understood. With this motivation, a proof-of-concept study for stabilizing the CEI formed on model $LiNi_xMn_yCo_zO_2$ (NMC/x+y+z=1) cathode was extensively studied. An in-depth investigation into reversible lithiation/delithiation at extremely high temperature (~100ºC) operation in thermally stable ionic liquid electrolyte combination was evaluated.[2] Even though ionic liquids are thermally stable, electrochemical interaction between reactive cathode surface and ionic liquid electrolyte is unfavorable. This presentation will delve into the fundamental understanding of this phenomenon. Further, the depth-dependent interfacial properties of the CEI formed on the NMC family cathodes cycled at high temperatures were understood using energy-tunable hard X-ray photoelectron spectroscopy (HAXPES). In addition, the bulk and surface electronic structure evolution at extreme temperature was probed using soft and hard X-ray absorption spectroscopy investigations. Stabilization of the reactive NMC cathode surface at extremely high temperatures using conformal surface passivation and layer-to-spinel structural transformations were visualized using high-resolution transmission electron microscopy (HRTEM) investigations (Figure 1). In this study, understanding the high-temperature interfacial stability of the NMC cathode materials through multimodal spectroscopy and microscopy was focused to transform the ambient temperature Li-ion battery technology to extreme temperature applications.

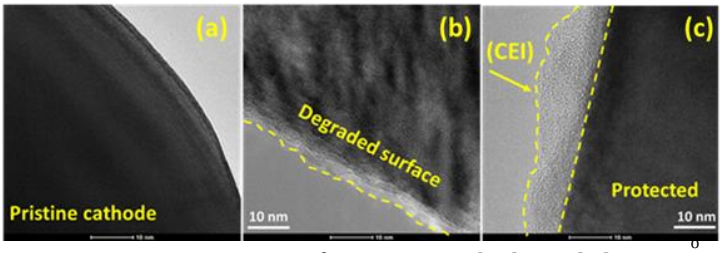

Figure 1. HRTEM images of NMC333 cathode cycled at 100 C with and without CEI protection. (a) Pristine NMC333 cathode powder, (b) Cycled cathode without film forming additives,(c) Cycled cathode with film forming additives

References:

1. Lin, X.*, et al.*, *Chemical Society Reviews* (2016) **45** (21), 5848
2. Nagarajan, S.*, et al.*, *Chemistry of Materials* (2022) **34** (10), 4587