Reduction of Full Cell Impedance by Dissolving Degradation Films

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It is well known that changes in the SEI can have deleterious effects on battery performance. However, other than a general presumption that thicker SEI films have higher impedances than thinner ones, little is known about how specific changes in SEI properties-chemical, physical, electrical, morphological-affect specific aspects of performance such as impedance. Even less is known about properties of degradation films on the positive electrode. For this work we take advantage of the fact that the degradation films on both electrodes can be dissolved, Figures 1 and 2, and that the amount of dissolution depends on both the solvent and its temperature. Importantly, not all of the species in the films dissolve at the same rate. In the example shown in Figure 2, a limited amount of soaking brings the Mn concentration at the surface of a degraded NCM electrode below that in the bulk, while the concentrations of Ni and Co remain above that in the bulk. Employing the isotope tracer approach that we developed previously [Electrochemistry Communications 13, 1035 (2011)], we use XPS and TOF-SIMS analyses to determine changes in the film thickness, composition, porosity depth profile, and Li⁺ diffusion rate with soaking. Relationships between changes in film properties and changes in battery performance are described. We show a significant reduction in impedance is possible from dissolving away these films.



Figure 1. <u>Anode</u>: Soaking at 60°C removes SEI on calendar aged cell.



Figure 2. <u>Cathode</u>: Li_2F^+ graph indicates removal of surface decomposition products. Mn/Co/Ni signals from the surface are different after soaking, but the bulk levels are similar. This shows that soaking removes surface species (metal fluorides, metal oxides, etc) containing Mn/Co/Ni.