NEW INSIGHTS INTO THE ELECTRODE/ELECTROLYTE INTERFACE ON POSITIVE ELECTRODES IN LI-ION BATTERIES

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Understanding and controlling the reactivity at the electrode/electrolyte interface (EEI) is one of the key issues for the development of high capacity and efficient lithium-ion batteries. The heterogeneous and partially catalytic reaction of the electrode with the electrolyte triggers the formation of surface films on the electrode surface which can cause degradation of the cell performance. Whereas the EEI layer properties are quite well known for negative electrodes such as lithium metal and graphite [1,2], the EEI layer on positive electrode materials is still puzzling. Especially the interface layers on high voltage and high capacity positive electrodes, whose potentials approach the limit of electrolyte stability against oxidation [3], is quite unexplored. One of the challenges in understanding the reactions at the surface of the electrode is the complicated composition of the positive electrodes, containing not only the active material but also conductive agents and polymeric binders, that can modify the EEI layers on the electrode. To bypass these ambiguities, there is a need for study model electrodes such as thin films or pure active material electrodes, which allow for investigating solely the reactivity of the electrolyte at the active material surface. Here, combining X-ray Photoelectron Spectroscopy (XPS and X-ray Absorption and Emission Spectroscopy (XAS/XES), of model electrodes, we will show how the species formed at the electrode/electrolyte interface are affected by change in charging potential and the structure and nature of the transition metal in the material. XES and XAS will be used to shed light on the change of electronic structure upon delithiation.

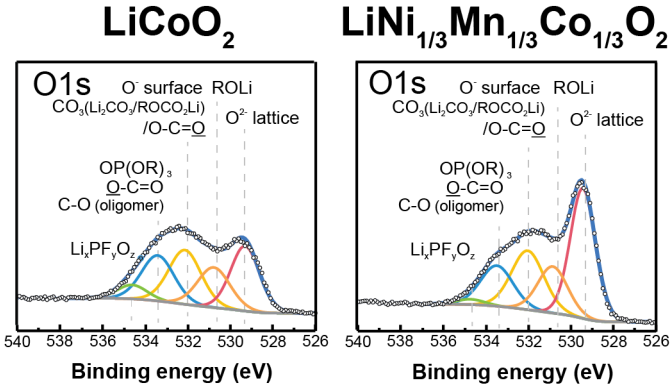

Figure 1: O1s XPS spectra photoemission lines for 100% $LiCoO_2$ or $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ electrodes charged at 4.6 V_{Li} at a rate of C/100 in 1M $LiPF_6$ 3:7 EC:EMC.

[1] Peled, E., *J. Electrochem. Soc.* **126,** 2047–2051 (1979).
[2] Aurbach, D. *et al.*, *J. Power Sources* **81–82,** 95–111 (1999).
[3] Xu, K. *et al.*, *Chem. Rev.*, **114**, 11503-11618 (2014).