Operando Characterization of Interfaces in Batteries

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Li-ion batteries are to dominate the high-end rechargeable energy storage market for a foreseeable future. Research and development of commercially relevant electrode/electrolyte systems focus today strongly on extending the lifetime of the cells. Progress is however hampered by the lack of fundamental understanding of the underlying lifetime-limiting processes. In the talk, progress on the development and application of characterization techniques for *operando* studies of electrochemical and chemical side reactions in next-generation Li-ion batteries will be presented.

Online electrochemical mass spectrometry (OEMS) provides unique means for mechanistic studies of side reactions in Li-ion batteries due to its unparalleled sensitivity and selectivity. Decomposition reactions involving (1) the electrode active material, (2) conductive carbon, (3) binder, as well as (4) the electrolyte solvent and (5) salt - along with (6) eventual additives - will be discussed. Particularly the critical role of Ni in governing the stability of Ni-rich layered mixed transition metal oxides ($LiNi_xCo_yMn_zO_2$) will be highlighted. Surface phase transformations during the initial cycles lead to composition- and potential-dependent evolution of O_2 under practical operation conditions (cf. figure below).[1] Electrochemically initiated decomposition of cell components initiates further secondary side reactions (e.g., as observed via the evolution of CO_2, HF, POF_3, etc.). The need for and strengths of complementary *operando* techniques such as Raman spectroscopy and electrochemical quartz crystal microbalance will be demonstrated.[2,3]

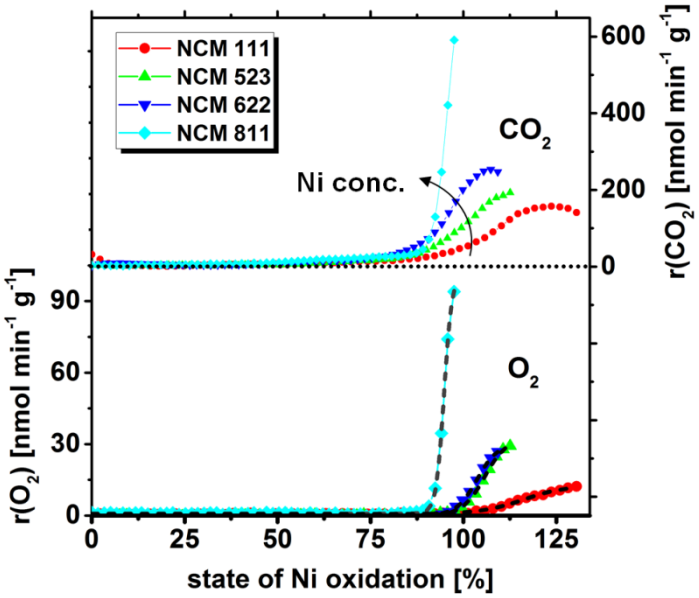

Figure 1. CO_2 and O_2 evolution from Ni-rich $LiNi_xCo_yMn_zO_2$ (NCM XYZ) during the 1st charge.[1]

References:
[1] D. Streich, C. Erk, A. Guéguen, F.F. Chesneau, E.J. Berg, J. Phys. Chem. C 121 (2017) 13481−13486.
[2] E. Flores, U. Aschauer, P. Novák, E.J. Berg, *submitted.*
[3] P. Kitz, P. Novák, E.J. Berg, *submitted.*