Clarification of the aging mechanisms of novel cathode-active materials via operando XAS/XES (operaXX)

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Public abstract

The increasing demand for efficient Li-ion battery materials with reduced environmental impact requests in-depth knowledge of the microscopic properties of the adopted compounds. Experimentally, x-ray spectroscopy is a powerful technique that enables monitoring the electrochemical charging cycle of batteries both ex situ as well as in operando condition. These measurements, however, need to be complemented by atomistic, quantum-mechanical simulations that are able to shed light on the involved mechanisms. Performing these simulations and understanding the electronic structure of battery materials is our task in the operaXX consortium. By means of density-functional theory embedded in a high-throughput computational screening framework for automatized calculations, we addressed the challenge of identifying the degradation products emerging from the delithiation process of battery materials composed of Ni-Co-Mn (NCM) as well as Ni-Co-Al (NCA) oxides in the last project phase. In the second phase which would be covered by this follow-up proposal, we will focus on the cathode materials themselves. Especially the delithiation process of these cathodes is of crucial interest. To compare with operando x-ray absorption spectra, we will utilize DFT-based projected density of states for delithiated phases at various concentration levels. However, due to the presence of different transition metals and the inherent complexity of these materials, determining their magnetization state poses a significant challenge, requiring substantial computational resources.