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Complementary Neutron-Based and X-Ray Measurements on the Lithiation Mechanism of LiAl Electrodes

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Metal alloys, such as LiAl, are gaining more interest as anode materials for lithium ion batteries because they exhibit a high theoretical capacity while being inexpensive. Aluminium has an almost three times higher specific capacity with 993 mAh/g in relation to the commonly used graphite anode. [1] During Lithiation, aluminium begins to form a solid solution with lithium, the so called alpha-LiAl phase. Once the solubility limit is reached upon lithiation, the alpha-LiAl undergoes a phase transition to the beta-LiAl phase. [2]

Al electrodes were electrochemically lithiated to different state of charges in coin cells with Li metal as counter electrode. X-ray diffraction measurements were conducted on the disassembled anodes with the goal to understand the lithiation mechanism and to determine the fractions of alpha- and beta-LiAl phases. The Rietveld refinements yield that the samples charged to SoC25 show a higher amount of alpha-LiAl. Simultaneously, the amount of beta-LiAl in the samples increases with higher SoC as expected.

Additional neutron depth profile measurements were performed in NPI CAS Rez at the CANAM infrastructure to determine the Li distribution throughout the lithiated samples. First results have confirmed that the lithiation of the aluminium starts at the surface where a higher Li concentration was identified. The higher charged sample also shows a stronger lithiation in the bulk of the Al anode. This shows that the whole anode is lithiated with a concentration gradient from surface to bulk material. In the lower charged sample, no Li was detected near the backside of the anode, indicating that pristine aluminium is there still present. Additional operando diffraction measurements are planned to further investigate the early stages of the lithiation and the nucleation of the beta-LiAl.

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