On the inhomogeneous nature of Li-intercalation and Li-plating on graphite-based anodes – an experimental study by means of high-resolution light microscopy under inert gas atmosphere

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Lithium-ion batteries (LIBs) currently represent the dominant technology in the field of rechargeable mobile and stationary energy systems. In order to ensure the longest possible life time, critical ageing mechanisms must be prevented. This includes, in particular, anode-side Li deposition ("Li plating"), which can lead to a rapid shortening of life time as well as to a deterioration of the safety characteristics [1]. Therefore, it is necessary to develop and validate a methodology for the identification and characterization of Li plating on todays widely used graphite anodes as well as for future Si/C composite anodes. Ex situ methods typically involve taking apart the cell to perform post mortem characterization. Since Li and LiC_6 are extremely reactive, ex situ techniques require very careful and resilient sample preparation workflows to minimize damage to the sensitive materials and maintain the cell conditions [2]. For example, LiC_6 reacts with moisture to form lithium (hydro) oxide. The reaction occurs by sequential formation of higher stages LiC_{12}, then LiC_{18} and then LiC_{24}[3]. Therefore, the cell de-assembling and the subsequent characterization has to be carried out under a controlled environment such as Ar or vacuum.

In this work, we want to present results from ex situ microscopic analysis of sensitive and highly reactive Li plating as well as lithiated graphite anodes. Based on high resolution surface imaging (i) the inhomogeneity of the lithium distribution, which can be recognized by distinct colors of lithiated graphite, along the electrode surface and within individual graphite particles is shown as a function of state of charge and charge rate. Also, (ii) Li plating, its onset, morphology and quantity as well as Li re-intercalation processes are studied – again depending on the state of charge and charge rate. For both cases (i and ii) the importance of a controlled inert gas environment and the influence of ambient air and moisture is investigated. Furthermore, we strive to deepen the understanding of the heterogeneous nature of Li plating and Li intercalation over the electrode volume of differently charged anodes by preparing cross sections with an Argon-polisher under inert atmosphere. To achieve these results, we set up a high-resolution light microscope in a glovebox and developed resilient preparation workflows and analytical routines in order to ensure a high reproducibility.

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