Investigations on Lithium Deposition as a Function of Current Densities with an Optically Accessible Cell

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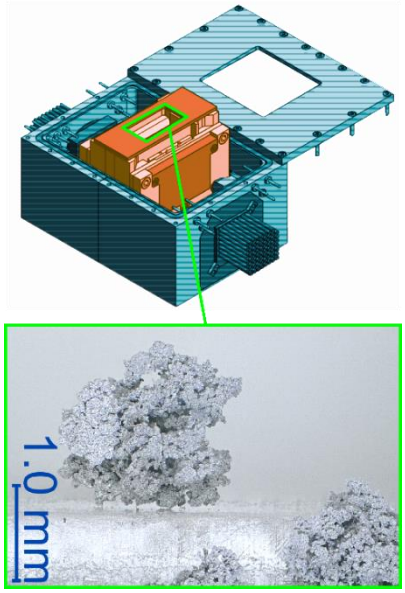
A market-ready rechargeable high-energy lithium metal battery (LMB) would be a revolution in the development of energy storage systems. The outstanding properties, such as a theoretical achievable capacity 10 times higher than graphite, a very low negative redox potential and a low atomic weight, make lithium metal an attractive anode material.[1,2] One of the main obstacles, which hinders the large-scale commercialization of LMB, is lithium deposition on metallic surfaces. Furthermore, growth of these deposition structures is difficult to predict and might lead to safety risks.[3] Hence, fundamental processes, such as site-specific deposition behavior, its characterization and prediction on lithium surfaces as a function of defined parameters, have to be investigated. One approach for such a study is to investigate the behavior of lithium metal deposition and stripping in an optically accessible in situ cell.[4-7]

In this work, a custom-designed in situ optical cell was used to investigate the lithium deposition behavior in a symmetrical lithium||lithium setup as a function of the current density. After a defined lithium-electrolyte contact time, an identical charge was transferred at various current densities (range from 0.05 mA cm^{-2} to 10 mA cm^{-2}). Electrochemical impedance spectroscopy (EIS) was conducted before and after each constant current (CC) phase. During the complete procedure in situ recordings were made with a digital microscope at a 5x magnification. After the electrochemical testing protocol was completed, close-up images from individual surface structures were captured at higher magnifications (50x – 250x).



Two regimes for lithium depositions were identified, including a transition region between them. Optical and electrochemical measurements show different results at lower and higher current densities. Trends for these regimes were identified based on the mean number of surface structures, characteristic length scales of structures, estimation of deposition volumes, overpotential as well as changes between EIS-measurements before and after the CC-phase as a function of current density. In the low current density regime, none to very few, larger lithium structures are observable. In the transition region, more and larger lithium structures are generated. Towards higher current densities, the number of structures increases exponentially while the dimensions decrease, approaching a threshold value. In context, these parameters allow, for example, conclusions on the electrode site dependency and porosity of the surface structures for the corresponding current densities.

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