Structuring thick high-energy silicon/graphite anodes by multilayer coating

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Lithium-ion battery cells with high specific capacity and energy density are one of the current research priorities in industry and science. The challenges of these so-called "thick" electrodes are transport limitations within the electrode: lithium ions cannot reach the deeper layers of the electrode coating, which leads to a drop in performance during charging and discharging. This phenomenon is a major hurdle in the further development of future high-capacity electrodes. One possible solution is to use multilayer electrodes with different layer properties. In this way, pathways for the Li-ions can be created that reduce the ionic resistance of the electrode and ensure an increase in fast charging capability.

The focus of this work is on two-layer anodes with different electrode layer configurations, whereby the influence of a varying areal capacity [4/6/8 mAh cm^{-1}] is specifically taken into account. For this purpose, an active material mixture of graphite, hard carbon and silicon is used (≈ 575 mAh g^{-1}). First, the processing of the different anode configurations is investigated, looking at the mechanical (adhesion), electrical (electrical resistance) and structural (porosity measurements) layer properties. A special focus is on characterization by means of impedance spectroscopy. Here, the electrodes are examined for their ionic conductivity and thus conclusions are drawn about the pore network and the tortuosity of the electrodes. In addition, promising multilayer anodes are tested for their electrochemical performance in full cells. The electrochemical characterization includes, in particular, a comprehensive evaluation of the charging performance (fast charging capability, Li-plating) in order to be able to evaluate the different layer configurations. With regard to the electrochemical characterization, it was found that the two-layer anodes with an active material mixture of all three materials in the upper layer showed a significant increase in ionic conductivity compared to a single-layer reference anode. In addition, the anodes also showed an increase in discharge capacity, especially at low C rates. One suggestion is that the addition of hard carbon could create a larger number of pores for the lithium ions, leading to improved ion transport and a reduction in transport limitations. In terms of areal capacity, it could be shown that the ionic conductivity decreases linearly with increasing areal capacity, which can be attributed to longer ion diffusion pathways.