Title: 3D printing of silicon-based anodes for lithium-ion batteries

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In order to meet the target for the next generation lithium-ion batteries, electrochemical performance such as energy and power density must be increased significantly at the same time. Optimized electrode architectures including 3D battery concepts and advanced materials are in development to achieve this goal. The use of silicon-graphite composite electrodes instead of graphite anodes is currently investigated. This is due to the fact that silicon can provide almost one order of magnitude higher specific energy density (3579 mAh/g) in comparison to natural graphite (330 - 372 mAh/g). However, during lithiation, i.e., lithium silicide formation, a volume expansion of about 300 % can take place, while during lithium intercalation in graphite about 10 % volume expansion can be observed. A huge volume expansion leads to a tremendous mechanical degradation of the anode resulting in a drop in capacity, and a limited battery lifetime.

In the presented study, laser induced forward transfer (LIFT) is applied as printing technology to develop sophisticated graphite and graphite-silicon electrode architectures with advanced electrochemical performances. LIFT was performed using a pulsed nanosecond UV laser with a repetition rate of up to 30 kHz and a maximal power of 10 W. To enable an accurate printing process during LIFT, the properties and compositions of the active material inks as well as the laser and process parameters have to be optimized. The printing process in combination with laser structuring provides a high flexibility regarding the final electrode design. In the presented study, the formation of multi-layer electrodes with spatial variation in electrode composition is achieved. As active materials silicon nanoparticles (SNPs) and various types of graphite, i.e., natural graphite, mesocarbon microbeads (MCMB), and artificial flake-like graphite with an average particle diameter of 1 µm up to 15 µm are utilized. The geometry and thickness of each printed layer is adapted with regard to an optimized electrochemical performance and cell lifetime. A single layer thickness of 5 µm up to 20 µm was achieved, while areal capacities of multi-layer anodes reaches values of 2 to 4 mAh/cm^2. In addition to the

applied active materials and architecture concepts, different solvent and binder systems are investigated with regard to process scalability and an improved environmental compatibility. The printed electrodes are electrochemically characterized by rate capability measurements at C-rates of up to 5C. A correlation between capacity retention and electrode architecture is achieved. The results are discussed in terms of upscaling and impact on the next generation anode material.