Evaluation of electrochemical performance tuning by laser structuring of electrodes and its impact on cell degradation mechanisms

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Abstract

Tuning of electrochemical properties of lithium-ion batteries by using ultrafast laser structuring of electrodes is a rather new technical approach. At Karlsruhe Institute of Technology (KIT) this technology was developed and established for the first time. However, quite recently, other research groups and institutions worldwide also recognized the high potential of this technology for lithium-ion battery production. Lately, a roll-to-roll laser processing infrastructure for structuring large footprint electrode materials for high capacity pouch cells and cylindrical cells was installed at KIT in order to push this technology to demonstrator level, namely technology readiness level 6. The high rate capability of the produced high energy battery cells were significantly improved and twice lifetime of cells could be achieved in comparison to cells with unstructured electrodes. In frame of research, cooperative, and industrial projects the impact of laser structuring for small and large footprint electrodes was studied with regard to lithium distribution caused electrochemical cycling. Hereby, different types of LiNi_xMn_yCo_{1-x-y}O₂ (NMC) as a cathode material and graphite as well as silicon-graphite as an anode material with areal capacities up to 4 mAh/cm² were investigated. Quantitative lithium distribution along entire electrodes were measured by laser-induced breakdown spectroscopy (LIBS). From the 3D elemental mapping starting points for electrochemical degradation could be identified. In cathode materials the local increase of lithium indicated the formation of electrical short cuts which were most related to electrode material inhomogeneity, e.g., by a macroscopic change in porosity or macroscopic film defects. The compressive stress applied to electrodes and separator during cycling has also a significant influence on lithium distribution and subsequent cell degradation. The appropriate type of laser generated patterns such as holes, lines or grids strongly depends on the application scenario as well as type of materials. For silicongraphite anodes the huge volume expansion during lithium silicide needs to be considered and more rectangular and broader structure features become relevant. Laser structuring of electrodes showed in all cases, for small and large footprint cathodes and anodes, a rather homogenized lithium distribution along the surface and electrode thickness. For full cells elemental mapping of electrodes facing each other was performed indicating the mutual influence on lithium distribution. It can be concluded that the laser-based 3D electrode concept is beneficial regarding a reduced cell degradation. In addition, it could be proven by LIBS that for the 3D battery concept new lithium diffusion pathways were generated which becomes activated at elevated C-rates. Those new lithium diffusion pathways counteract the increase of diffusion overpotential leading to a higher capacities also for high power operation.

Keywords: lithium nickel manganese cobalt oxide; silicon-graphite, laser structuring; 3D battery; lithiumion battery; laser-induced breakdown spectroscopy; electrode engineering, electrode manufacturing