Z03-3284 - *(Invited)* Elucidating the Hierarchical Structuring of Ultra-Thick-Three-Dimensional-Designed Electrodes for Lithium-Ion Batteries

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Abstract

The transition to sustainable energy systems requires lithium-ion batteries capable of delivering high-energy density, high-power output, and extended cycle life while maintaining cost-effective manufacturing. Current limitations arise from existing trade-offs between energy and power density in conventional two-dimensional (2D) electrodes. This study addresses these barriers by developing a scalable high-power GHz laser ablation process to fabricate three-dimensional (3D) microstructured electrodes, demonstrating enhanced electrochemical performance through optimized patterning parameters.

Conventional electrode architectures struggle to eliminate the competing demands of energy storage capacity and charge/discharge rates due to lithium-ion diffusion limitations in thick films, i.e. high-mass-loading configurations. While material innovations improve the properties, overall gains require electrode designs that accelerate and homogenize electrode wetting and shorten lithium-ion diffusion paths. Existing laser ablation techniques using nanosecond to femtosecond pulses in kHz up to MHz regimes face scalability constraints from low throughput and thermal damage risks. Especially for cathodes using water-based binder systems, a deeper understanding of laser ablation is necessary.

Therefore, in this study, electrodes were prepared using a water-based binder system with sodium carboxymethyl cellulose (Na-CMC) for both cathodes and anodes. The cathode contains lithium nickel manganese cobalt oxide (NMC 811) as active material, while artificial graphite was used in the anodes. Both electrodes were designed with ultra-high areal capacities, having around 8 mAh/cm^2 and up to 10 mAh/cm^2 for cathodes and anodes, respectively. As a result, an N/P-ratio of around 1.1 was achieved, considering an N/P ratio of around 1.1 and the different mass losses of both electrodes due to laser structuring.

This work is among the first to explore GHz burst-mode laser structuring of NMC 811 cathodes and graphite anodes with aqueous binders, systematically evaluating the interplay between laser pulse train duration (30 – 500 ns), laser repetition rate (100 kHz – 1 MHz), average laser power (1 – 100 W), and laser scanning speed (up to 20 m/s). The results of the individual materials are also compared to illustrate the different laser ablation characteristics. By operating in the GHz frequency domain, the technique achieves precise material removal through cumulative sub-threshold energy pulses, minimizing the heat-affected zone while enabling a significant boost in processing rates.

Optical and electron microscopy revealed that material dependent variations of burst lengths produced optimal channel geometries. Electrochemical analyses using full-cell designs with 3D-structured electrodes demonstrated an enhancement in capacity retention for high charge and discharge rates compared to cells with unstructured electrodes, in addition to an increase in volumetric energy density due to the enhanced electrode compaction.

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