Abstract

The increasing demand for efficient and sustainable energy storage systems, driven by the electrification of transportation and the integration of renewable energy sources, has positioned lithium-ion batteries (LIBs) as a key enabling technology. Despite their technological maturity, current LIBs face limitations in terms of energy density, safety, and environmental sustainability, particularly due to the use of critical raw materials and the challenges associated with end-of-life management. In this context, advanced processing strategies represent a complementary pathway to materials development, offering the possibility to tailor interfaces, improve performance, and enhance sustainability.

This dissertation investigates the role of laser-based processing techniques for the development of high-performance and sustainable lithium-based battery electrodes. The research activity is structured around three main directions: (i) engineering of current collectors for anode-less lithium metal batteries (ALLMBs), (ii) laser structuring and surface modification of battery electrodes, and (iii) direct recycling of high-energy cathode materials.

The first part of the work focuses on the laser-induced surface modification and oxidation of copper current collectors for ALLMBs. These systems, which rely on in situ lithium plating, offer the potential for significantly increased energy density but are strongly limited by interfacial instability and low Coulombic efficiency. Laser processing is employed to modify both the surface morphology and chemistry of copper, promoting more uniform lithium nucleation and growth. The results show a reduction in nucleation overpotential, improved reversibility of lithium plating and stripping, and enhanced cycling stability compared to untreated current collectors.

The second part of the dissertation addresses the use of laser-based techniques for electrode structuring and current collector texturing. Laser structuring is applied to LNMO cathodes and Si/C anodes to introduce controlled architectures aimed at improving ion transport and reaction kinetics. The structured electrodes exhibit enhanced rate capability and improved performance, particularly at higher thicknesses where diffusion limitations are more pronounced. In parallel, laser texturing of aluminum current collectors is investigated to improve adhesion between the active material layer and the substrate. The modified surfaces lead to increased interfacial strength and improved electrochemical stability over extended cycling.

The final part of the work is dedicated to the development of a direct recycling approach for spent NMC811 cathodes. A delamination process based on propylene carbonate is combined with a microwave-assisted relithiation treatment to regenerate the active material. This method enables the recovery of cathode materials while preserving their morphology and structural integrity. The regenerated materials show electrochemical performance comparable to that of pristine NMC811, demonstrating the effectiveness of the proposed approach.

Overall, this work highlights the potential of laser-based processing as a versatile and scalable tool for battery manufacturing and refurbishment. By enabling precise control over surface properties and electrode architecture, these techniques contribute to improving electrochemical performance and addressing key challenges related to next-generation battery technologies and circular economy strategies.