Abstract

This doctoral research investigates the development of sustainable lithium-ion battery (LIB) anodes by integrating electrochemical performance, electrode manufacturing processes, and environmental impact assessment, driven by the need for high-energy-density storage technologies with reduced environmental impact of battery production.

The study explores the transition from conventional graphite to silicon-based anodes, including silicon-composite (~10% Si) and silicon-dominant (~80% Si) systems. Although silicon offers a significantly higher theoretical capacity, its implementation is limited by large volume changes during cycling, causing mechanical degradation, loss of electrode cohesion, and unstable electrochemical performance. To address these challenges, electrode formulation was optimized using aqueous binder systems and natural polysaccharides.

Slurry mixing, coating, and drying were systematically optimized at laboratory scale by evaluating different binders and processing conditions, while pilot-line activities were carried out at a preliminary level to assess process transferability. Graphite electrodes exhibited stable slurry properties and uniform coatings at both laboratory and pilot scale, confirming the robustness of conventional CMC/SBR systems. The introduction of silicon (10 wt% Si) increased sensitivity to formulation parameters, leading to slurry instability and coating inhomogeneity, further amplified at pilot scale. These effects became critical for silicon-dominant electrodes (80 wt% Si), where insufficient cohesion and poor adhesion to the current collector strongly affected electrode integrity and processability at pilot scale. Increasing silicon content therefore shifts the main challenges from electrochemical performance to processability and manufacturing stability.

Binder chemistry was identified as a key factor: while CMC/SBR systems showed limitations for silicon-based electrodes, PAA-based formulations improved active material dispersion, particle cohesion and adhesion at the current collector interface, and electrochemical performance, particularly for silicon-dominant systems. Natural polysaccharides, introduced at 1 wt% in the formulation, further influenced slurry rheology and coating quality. In addition, laser texturing of the copper current collector, particularly DLIP (Direct Laser Interference Patterning)-treated surfaces, enhanced electrode performance, highlighting the role of electrode–current collector interface.

A cradle-to-gate Life Cycle Assessment (LCA) based on primary experimental data showed that environmental impacts depend not only on material composition but also on process-related factors, especially energy-intensive steps such as drying. Per unit of capacity (1 Ah), silicon-based anodes reduced carbon footprint (kg CO_2-Eq) compared to graphite by ~40% for silicon-composite and up to ~97% for silicon-dominant systems. These benefits, however, depend on the selected functional unit and system boundaries. A sensitivity analysis comparing a global supply chain scenario, representative of current industrial conditions, with a European-based scenario highlighted the influence of electricity mix and transportation. A preliminary extension to the full-cell level evaluated the translation of these improvements at system level.

Overall, the work shows that advanced LIB anodes require an integrated approach in which material selection, manufacturing processes, and sustainability are addressed simultaneously, with silicon-based materials playing a key role in enabling next-generation high-energy-density systems. The results of this study provide a basis for the future development of design-support tools in which material selection, electrode formulation, and process parameters are directly linked to both electrochemical performance and environmental impact.