Materials Science and Engineering Dissertation Defense

Sustainable Solutions for Lithium–Based Rechargeable Batteries

School for Engineering of Matter, Transport and Energy

Yuhui An

Advisor: Yoon Hwa

Abstract

Lithium-based rechargeable batteries represent a critical energy storage platform, yet their conventional lifecycle is hindered by severe environmental and economic challenges. The heavy reliance on toxic solvents and persistent polymer binders inflates fabrication costs and complicates circular recovery of end-of-life cells. To establish a more sustainable trajectory, two critical advancements were developed, spanning from cathode fabrication to end-of-life anode recycling.

First, a scalable, solvent- and polymer-binder-free manufacturing method for lithium-sulfur (Li||S) positive electrodes was established via thermal-assisted dry pressing, reducing fabrication costs by a factor of 2.1 relative to conventional slurry casting. By leveraging sulfur as a thermally activated structural binder at 80 °C, this approach eliminates hazardous solvents and inactive polymers entirely. A critical factor is the temperature-driven softening and rheological transformation of sulfur, enabling it to flow and function as an effective interparticle adhesive. To fully optimize this consolidation mechanism, the intraparticle distribution of sulfur within sulfur-carbon composites was systematically investigated using two model systems: a ball-milled composite (B-S/C) with surface-aggregated sulfur and a melt-diffusion composite (T-S/C) with uniformly confined sulfur. Under applied thermal stress, B-S/C provided high initial cohesion but formed crack-prone crusts and sulfur-depleted rims through heterogeneous consolidation. Conversely, T-S/C enabled cohesive interparticle fusion while preserving a percolated porous framework. Peel testing and ex situ SEM/EDS confirmed that T-S/C maintained superior mechanical integrity with smooth, continuous morphology, whereas B-S/C suffered bulk particle detachment and pronounced cracking. The optimized T-S/C architecture delivered a reversible capacity of 932 mAh g^{-1} after 500 cycles at 1.0C in coin cells.

Second, an electrode-resolved, reagent-minimized recycling route recovered high-value graphite and copper current collectors from spent anodes. Conductivity-guided aqueous pre-rinsing, rapid thermal processing, and surfactant-free high-shear delamination achieved over 97% graphite recovery while fully preserving copper foil integrity. Subsequent boric-acid and heat treatments cleared residual contaminants and restored structural ordering. The regenerated graphite outperformed froth-flotation graphite, and a full cell with an LFP cathode retained 107.2 mAh g^{-1} after 300 cycles at 0.2C. Collectively, these advances provide a closed-loop framework for the sustainable manufacturing and recycling of high-performance lithium-based batteries.

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