Engineered Current Collector Interface for High Energy Density Li-Ion Batteries

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Abstract: Resistive interfaces within the electrodes limit the energy and power densities of a battery, e.g., a Li-ion battery (LIB). Typically, active materials are mixed with conductive additives in organic solvents to form a slurry, which is then coated on current collectors (e.g., bare or carbon-coated Al foils), to reduce the inherent resistance of the active material. Although many approaches using nanomaterials to either replace Al foils or improve conductivity within the active materials have been previously demonstrated, the resistance at the current collector-active material interface (CCAMI) - a *key factor for enhancing the energy and power densities* remains unaddressed. We show that carbon nanotubes (CNTs), either directly grown or spray coated on Al foils, are highly effective in reducing the CCAMI resistance of traditional LIB cathode materials ($LiFePO_4$ or LFP and $LiNi_{0.33}Co_{0.33}Mn_{0.33}O_2$ or NMC). Moreover, the CNT coatings displace the need for currently used toxic organic solvents (e.g., N-Methyl-2-pyrrolidone or NMP) by providing capillary channels, which improve the wetting of aqueous dispersions containing active materials. The vertically aligned CNT-coated electrodes exhibited energy densities as high as: 1) ~500 Wh kg^{-1} at ~170 W kg^{-1} for LFP, and 2) ~760 Wh kg^{-1} at ~570 W kg^{-1} for NMC. The LIBs with CCAMI-engineered electrodes withstood discharge rates as high as 600 mA g^{-1} for 500 cycles in case of LFP, where commercial electrodes failed. The CNT-based CCAMI engineering approach is versatile with wide applicability to improve the performance of even textured active materials for both cathodes and anodes.