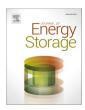
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A simple formula to fabricate high performance lithium metal capacitors

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ABSTRACT

Energy storage technologies that are low-cost, with long cyclability, high rate-capability as well as high energy and power densities are under intensive investigation for sustainable clean energy transition. In this paper, we report a high-performance lithium metal capacitor (LMC) achieved by a simple slurry modification during the cathode film preparation. We show that a mere substitution of \sim 0.4 wt% conductive carbon by single walled carbon nanotubes (SWCNTs) increased the specific energy of LMCs by 22 %. Porous carbon cathode in this study was obtained from a non-edible biomass (coconut rachis); the optimized sample showed desirable surface characteristics (surface area \sim 1933 m² g⁻¹ and pore diameter \sim 2.0 nm) as well as high edge-plane fraction (ratio between relative density of edge and basal plane \sim 0.4). Cathode with no SWCNTs show a specific capacitance (C₅) of ~133 F g⁻¹@0.1 A g⁻¹ in the potential window 2.0–4.0 V in the Li//LiPF₆//AC device configuration. Removal of conductive carbon by SWCNTs up to ~0.6 wt% increased electrical conductivity of the cathode; however, the charge storability enhancements were only up to \sim 0.4 wt%. The optimum device delivered a C_S \sim 188 F·g⁻¹@100 mA·g⁻¹ in the potential window 2.0–4.0 V with improved rate capability and cycling stability. Electrochemical impedance spectroscopy was used as a tool to understand the charge kinetics at the electrode; these studies collectively validated the observed enhancements in the charge storability. The device hereby developed showed superior specific capacity than most of the reported lithium-ion capacitors and comparable to some of the LMBs. The perceived 22 % increase in the specific energy by a mere 0.4 wt% SWCNT substitution is a step forward in fabricating the high-performance LMCs in addition to support the sustainability agenda through the carbon-negative precursors.

1. Introduction

The Li-ion batteries (LIBs) are currently dominating the energy storage market; however, higher energy and power density requirements to widen the electrification of the transport sector fuel the research on alternate devices such as lithium metal batteries (LMBs) and lithium metal capacitors (LMCs). The graphite anode in commercial LIBs have lower specific capacity (\sim 372 mAh·g⁻¹) and relatively high density (\sim 2.26 g·cm⁻³) whereas lithium metal anode in LMBs and LMCs have high specific capacity (\sim 3860 mAh·g⁻¹) and several-fold lower density (\sim 0.59 g·cm⁻³) allowing the latter to be high performing lightweight devices [1,2]. The LMBs and LIBs share similar specially

prepared multicomponent metal oxide cathodes made using energy and materials intensive processes, leading to high cost per unit energy storage (~\$100/kWh) in them [3]. On the other hand, LMCs use porous carbon electrodes due to their desirable surface, electrical, and electrochemical properties, which could be obtained from carbon negative precursors such as biomass, thereby making them potentially lower cost. However, their performance is not on a par with their battery analogues because of the charge storability limitations.

Renewable porous carbon extracted from biomass had gained tremendous attention owing to their high abundancy. Worldwide coconut production increased steadily from 51.19 million tons in 2000 to 63.68 million tons in 2021 [4]. Even though the fruits have numerous

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