Thesis Title:

Novel Carbon Architectures for Large-Sized Aqueous Supercapacitors with Wide Potential Window

ABSTRACT

Supercapacitors are high power electrochemical energy storage devices, known for high power delivery capability, long cycle life, high safety and ease of fabrication. However, the large-scale development of supercapacitor technologies is often limited by challenges related to electrode material scalability, electrolyte safety, narrow operating voltage windows and environmental footprint. Several laboratory-scale studies report high electrochemical performances at various cell configurations. However, translation to industrially viable devices remain a major bottleneck. Carbon-based materials are widely employed as supercapacitor electrodes. In this work, the design and development of scalable carbon architectures for large-sized aqueous supercapacitors, with wide operating voltage window, is systematically investigated. Three distinct carbon morphologies viz., bowl-like carbon structures (BCS), capped carbon structures (CCS) and reinforced carbon nanofibers (R-CNFs) are synthesized. Amongst these, electrospun R-CNFs shows the potential to be the most suitable candidate due to their ease of scalability, high yield, reproducibility, mechanical integrity and ease of scale-up.

The electrochemical performance of R-CNFs is comprehensively studied in both aqueous and non-aqueous electrolyte systems using three-electrode and symmetric device configurations. A moderately concentrated aqueous electrolyte (5 M $NaClO_4$ in De-ionized water) is able to deliver a wide electrochemical stability window without employing water-in-salt (WiS) electrolytes. A stable operating voltage window of up to 2.3 V is achieved in aqueous systems with electrochemical performance comparable to non-aqueous (1 M $NaClO_4$ in EC:PC

(1:1 v/v)) counterparts. Proving the industrial viability of aqueous electrolyte by offering advantages in safety, cost and environmental impact. Life cycle assessment studies further confirm the significantly lower carbon footprint of the aqueous-based devices.

The optimized electrode-electrolyte system is successfully tested in a large-area pouch cell supercapacitors (LSSCs) of dimensions up to (10×15) cm^2. The device demonstrates excellent energy and power densities, along with long-term cycling stability exceeding 25000 cycles. In addition, a gel-based polymer composite separator is developed to address electrolyte leakage and mechanical stability issues, enabling further widening of the operating voltage window.

Keywords: Supercapacitor, reinforced carbon nanofibers, aqueous electrolyte, large-area device, scalability, energy storage, gel separator