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Integrating the multifunction necessary for electrochemical energy storage into energy- and size-scalable ultraporous nanoarchitectures¹

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Designing high performance energy-storage devices that combine nanometric feature size with well-wired transport paths requires an architectural perspective. We chose carbon aerogel-like nanofoam papers as attractive plug-and-play electrode substrates because of such desirable properties as high specific surface area, electronic conductivity, and through-connected pore structure. Achieving this blend of desirable properties requires an optimal balance of critical architectural features: (1) open, 3D interconnected macropores sized at 100 to 300 nm (a difficult-to-obtain size range in porous carbons) and (2) pore walls of a size that reduce dead weight and volume (preferably ca. 20-nm wall thickness for 100- to 300-nm voids), yet retain mechanical strength and flexibility without compromising electronic conductivity (preferably ca. 20 S/cm). Carbon nanofoam papers provide a low cost and scalable nanocomposite that exists within this “Goldilocks zone” of desirable properties and which has catalyzed breakthroughs in our work with asymmetric electrochemical capacitors, air cathodes for metal/air batteries, lithium-ion batteries, 3D batteries, and semifuel cells. New charge-storage or catalytic functionality is imparted to internal carbon walls simply by transporting reactants within the 3D macroporous. Self-limiting modification strategies allow us to incorporate conformal, nanoscopic “paints” of metal (Mn, Ti, Ru, Fe) or polymer (redox-active or electron insulating) or to specifically adsorb metal nanoparticles (Pt, Au, Pd, Ag) throughout the macroscopic thickness (0.07 to 0.3 mm) of carbon nanofoam papers as dictated by the requirements of a specific end application. For instance, modification with 10-nm MnO_x increases the mass-, geometric-, and volume-normalized capacitance (2- to 10-fold) relative to the native carbon nanofoam without significantly altering its high-rate character and provides a structure that can be used in an asymmetric electrochemical capacitor or used to an air cathode in a Zn/air cell to electrocatalyze oxygen reduction and provide pulse power. Our redesigned carbon nanofoam offers a versatile design platform for much-needed advances in a broad range of multifunctional energy storage and conversion.

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