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**Controlling Electric Double-Layer Capacitance and Pseudocapacitance in Heteroatom-Doped Carbons Derived from Hypercrosslinked Microporous Polymers**

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Abstract: Hypercrosslinked polymers (HCPs) are an important class of porous materials that can be synthesized from aromatic precursors using a one-step “knitting” procedure. This scalable process allows wide synthetic diversity and ease of functionalization. However, pristine HCPs lack electrical conductivity, which limits their potential for electrochemical applications. Supercapacitors are energy storage devices with advantages over conventional batteries such as high power densities, rapid charge speeds, and superior cyclability. In this work, carbonization of functionalized HCPs yields highly conductive and porous materials that can be used as supercapacitor electrodes. Both electric double-layer capacitance (EDLC) and pseudocapacitance (PC) mechanisms are observed. The relative EDLC and PC contributions were quantified for a range of 20 HCP-derived materials, thus allowing a controlled approach to tuning the energy storage properties. The HCP-based carbons show ideal supercapacitor behavior and the best performing material, which shows 63% PC, displays exceptionally high capacitances of up to  $374 \text{ F g}^{-1}$ , excellent capacitance retention at fast charging speeds, and stability for up to 15,000 charge/discharge cycles.

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