GRAPHENE AEROGELS FOR HIGH-PERFORMANCE ELECTROCHEMICAL ENERGY STORAGE

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Developing three-dimensional (3D) graphene assemblies with properties similar to those individual graphene sheets is a promising strategy for graphene-based electrode materials . Typically, the synthesis of 3D graphene assemblies relies on van der Waals forces for holding the graphene sheets together, resulting in bulk properties that do not reflect those reported for individual graphene sheets. Here, we present the use of sol- gel chemistry to introduce chemical bonding between reduced graphene oxide sheets to yield ultra-low-density three-dimensional macroassemblies of graphene sheets that exhibit high electrical conductivities and large internal surface areas. These materials are prepared as monolithic solids from suspensions of single-layer graphene oxide in which organic sol-gel chemistry is used to cross-link the individual sheets. The resulting gels are supercritically- or freeze-dried and then thermally reduced to yield graphene aerogels with densities approaching 10 mg/cc. Adjusting synthetic parameters allows a wide range of control over surface area, pore volume, and pore size, as well as the nature of the chemical cross-links (sp2 vs sp3). These 3D graphene materials exhibit an improvement in bulk electrical conductivity of more than 2 orders of magnitude (∼100 S/m) compared to graphene assemblies with physical cross-links alone (∼0.5 S/m). The graphene aerogels also possess large surface areas (~600 m^2/g) and pore volumes (~3 cc/g), making these materials viable candidates for use in capacitive energy storage, catalysis, and sensing applications. Overall, the bulk properties of the graphene-based aerogels represent a significant step toward realizing the properties of individual graphene sheets in a 3D assembly with surface areas approaching the theoretical value of an individual sheet.