

Thermoelectric Properties of Graphene-Carbon Nanotube Aerogels as ‘Organic’ Energy Harvesters

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Abstract

We prepared graphene-based three-dimensional scaffolds comprising different ratios of graphene oxide (GO) and carbon nanotubes (GA-CNT, hereon) using facile hydrothermal technique. The resulting hydrogels are freeze-dried and thermally treated to yield structurally ordered aerogels with ultralow densities and tunable mesoscopic pore sizes. These ‘all carbon’ aerogels prepared as monolithic solids from suspensions of GO nanosheets and small diameter multiwalled and single-walled carbon nanotubes in which organic wet chemistry is used to cross-link the components for multiplex hierarchical topologies. In contrast to methods that utilize physical cross-links between GO nanosheets, this approach with polymeric linkers and organic functionalization provides covalent carbon bonding among the graphene nanosheets and molecular attachment with carbon nanotubes, respectively, facilitating rapid and facile electron or ion transport. As a result they exhibit highly interconnected network topology with large internal surface areas thus promoting enhanced surface ion adsorption which makes these mesoporous materials viable candidates for use in thermal and thermo-electrochemical energy harvesting and energy storage technologies. They have shown improved electrical conductivities ($> 5 \text{ S.cm}^{-1}$), Seebeck coefficient ($> 0.5 \text{ V.K}^{-1}$), and moderate thermal conductivity ($\sim 0.028 \text{ W.m}^{-1}.\text{K}^{-1}$). We also used complementary characterization techniques including electron microscopy, Raman spectroscopy and scanning electrochemical microscopy to establish *microstructure-processing-property-performance* correlations.