

3D Printing of Ultra-Compressible, Highly Conductive Graphene Aerogels

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Abstract

Graphene aerogels are typically micro- and mesoporous (pores <50 nm), ultra-lightweight, conductive materials that can achieve surface areas in excess of 1000 m²/g.^{1, 2} As such, they are used in a wide range of applications ranging including catalysts and catalyst supports,^{3, 4} energy storage and conversion,⁵ and sorbents for water purification.⁶ Aerogels are made via the sol-gel process, in which a reaction solution is gelled and the solvent is extracted in such a way as to leave the porous solid matrix intact.⁷ Though their pore sizes can typically be tuned by varying the synthetic parameters of the sol-gel process,^{8, 9} limitations do exist. For example, the tortuous, random porous network of graphene aerogels can significantly affect supercapacitor performance by limiting ion and electron transport. Thus the fabrication of a graphene aerogel with tailored macro-architectures to facilitate mass transport via a controllable and scalable assembly method remains a significant challenge.

Here, we report the fabrication of periodic graphene aerogel microlattices for supercapacitor applications, via a 3D printing technique known as direct-ink writing (DIW).¹⁰ These novel graphene structures possess an unprecedented, synthetically controlled hierarchical pore network that spans several orders of magnitude (nanometers to millimeters). (**Figure 1**) The key factors in developing these 3D graphene electrodes were developing an extrudable graphene-oxide (GO) based ink, and modifying the 3D printing method to accommodate aerogel processing, specifically “wet-printing” instead of the traditional dry printing process.¹¹ The 3D-printed graphene aerogel electrodes are lightweight, highly conductive and exhibit excellent electrochemical properties. In particular, the supercapacitor devices using the 3D-printed electrodes with thicknesses on the order of millimeters, display capacitance retention (~90% at 10 A/g) and power densities (>4 kW/kg based on full device mass), performance which equals or exceeds that of reported devices made with electrodes 10-100 times thinner. These results highlight how the 3D printing of novel materials, such as graphene aerogels, can significantly expand the design space for fabricating complex, high performance energy storage devices that can be optimized for a broad range of applications.

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Figures

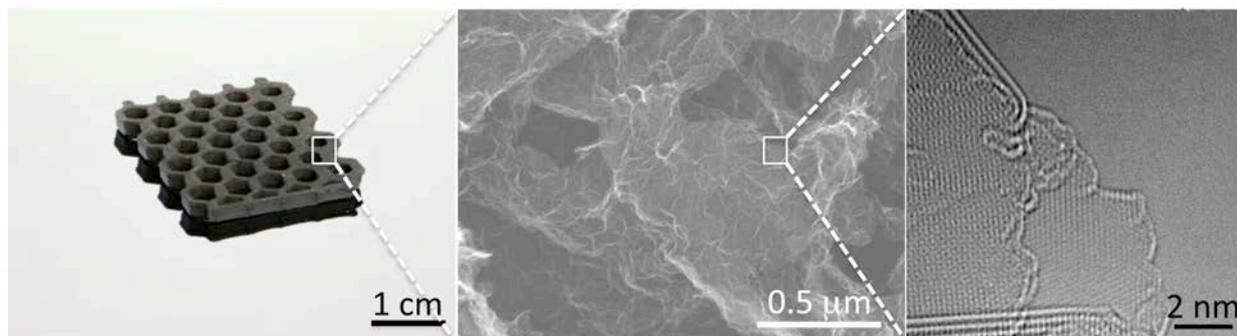


Figure 1. Images of 3D-printed graphene aerogel honeycomb lattice taken at various levels of magnification using optical (left), scanning electron (middle), and transmission electron (right) microscopy.