Simultaneous Sheet Cross-Linking and Deoxygenation in the Graphene Oxide Sol-Gel Transition: Supporting Information

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Nitrogen Porosimetry

Gravimetric surface area and pore volume were determined experimentally using a Micromeritics ASAP 2020 porosimeter. Samples were degassed overnight under vacuum at 150 °C, and then an isotherm was collected for relative pressures from $P/P_o = 0.06 - 1.00$, where P_o is the saturation pressure of nitrogen at 77K, ~760 mmHg (Figure S1). Mesopore surface area was calculated from the low-pressure ($P/P_o = 0.07 - 0.20$) adsorption data by the Brunauer-Emmett-Teller (BET) model.¹ Pore volume was obtained by applying the Barrett-Joyner-Halenda (BJH) model to the adsorption branch of the isotherm.² Another isotherm was then obtained for lower pressures: $P/P_o = 3x10^{-6} - 0.03$ (data not shown), and the BET model was used to obtain the micropore surface area. Micropore volume was calculated using density functional theory (DFT) modeling of slit-shaped pores.³ Values of surface area and pore volume are listed in Table S1.

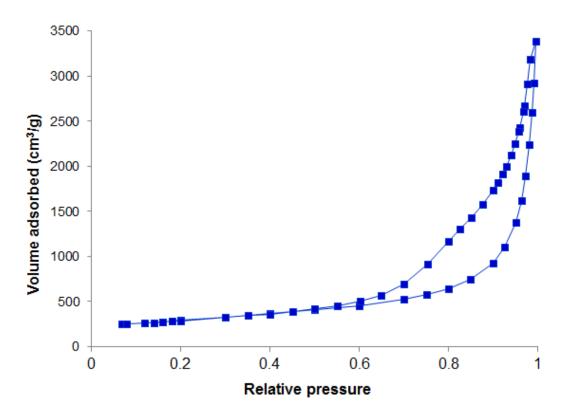


Figure S1. Nitrogen adsorption isotherm for the fully reacted GO gel

BET surface area (mesopore analysis)	1019.3 m ² /g
BET Surface Area (micropore analysis)	1141.8 m ² /g
BJH adsorption pore volume (17-3000 Å)	5.24 cm ³ /g
Micropore volume (≤ 29.49 Å)	0.38 cm ³ /g

Table S1. Surface area and pore volume values from nitrogen porosimetry analysis

Raman Spectroscopy

Raman spectra were taken with a Renishaw inVia microscope using the 514 nm line of an Ar ion laser. Gelling appears to have no significant effect on the Raman spectrum. In Figure S2, the spectrum for GO overlaps almost perfectly with the spectrum of the as-made aerogel. Annealing at 1050 °C increases the I_D/I_G ratio from 0.83 to 0.95, but the rest of the spectrum remains identical. The increase in I_D/I_G ratio is due to a departure from the Tuinstra-Koenig relation at high levels of disorder; increasing the size of sp² domains actually enhances the height of the D peak for nanocrystalline domains.⁴

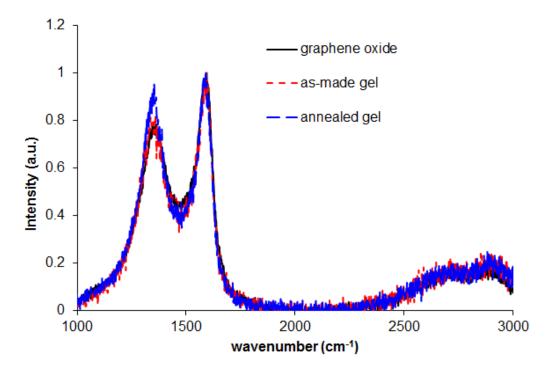


Figure S2. Raman spectra of graphene oxide, the as-made aerogel, and the aerogel after annealing. Spectra are background subtracted and normalized to the G peak.

References

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