-- Supporting Information --

Bulk Preparation of Holey Graphene via Controlled Catalytic Oxidation

Yi Lin,*^{*a*} Kent A. Watson,^{*a*} Jae-Woo Kim,^{*a*} David W. Baggett,^{*b*} Dennis C. Working^{*b*} and John W. Connell^{*b*}

^a National Institute of Aerospace, 100 Exploration Way, Hampton, VA 23666-6147, USA. Tel: 1 757 864 2219; ^b Mail Stop 226, Advanced Materials and Processing Branch, NASA Langley Research Center, Hampton, VA 23681-2199, USA ^{*}E-mail: yi.lin@nianet.org



Figure S1. An SEM image of a $(Ag-G)_{10}$ sample subjected to thermal treatment at 300 °C in air for 10 h. Very long and irregular Ag-etched tracks can be seen; most catalytic Ag nanoparticles in this region had probably dissociated from the graphene sheet.



Figure S2. Size distributions of the holes in (a) a hG₁ sample (4.9 ± 1.9 nm) and (b) a hG₁₀ sample (21.5 ± 31.5 nm) – the same samples shown in Figure 6. Inset of (b) showed the size distribution plot of the hG₁₀ sample prepared in larger scale (18.2 ± 20.2 nm; corresponding to the same sample shown in Figure 9a).



Figure S3. SEM images of (a) a hG_1 sample and (b) a hG_{10} sample for which Step-II thermal treatments were both carried out at 350 °C in air for 3 h.



Figure S4. DTG curves (air, 5.4 $^{\circ}$ C/min) of the hG₁₀ and hG₁ samples in comparison with the starting graphene sample.



Figure S5. Preliminary electrochemical evaluations: (a) cyclic voltammetry curves of a hG_{10} electrode at scanning rates from 10 (most inner curve) to 500 mV s⁻¹ (most outer curve). (b) specific capacitance values of hG_{10} in comparison with those of a control graphene sample (with 2 h 2.6M nitric acid reflux only). The measurements were carried out in 0.5 M Na₂SO₄ using three electrode configuration with Pt wire as the counter electrode and Ag/AgCl as the reference electrode. The working electrode was prepared by deposition of a drop of DMF dispersion of hG_{10} (~15 µg) onto a conductive ITO slide (negligible capacitance) followed by drying in air.