Experimental section

Synthetic Procedure

The graphene oxides (GO) were prepared from natural graphite using the Hummers' method with minor modifications.[S1] 0.5, 1, 2, and 4 grams of ZnO nanoparticles (Sigma-Aldrich) were added to 20 mL of GO solution (1 mg mL^{-1}), to generate the HGN-0.5, HGN-1, HGN-2, and HGN-4 samples, respectively. After sonication for 12 h, the mixed solution was filtered with a polytetrafluoroethylene (PTFE) membrane. The obtained ZnO/GO film was transferred into a horizontal furnace and calcined at 900 °C under nitrogen atmosphere for 4 h, using a ramp rate of 3 °C min^{-1}. After cooling the sample down to room temperature, holey graphene materials were obtained. These samples were referred as HGN-*X*, where *X* represents the amount of ZnO used in the reaction.

Structural Characterization

The scanning electron microscopy (SEM) observation were performed on a Hitachi SU-8000 field-emission scanning electron microscope. Transmission electron microscopy (TEM), scanning transmission electron microscopy (STEM) and energy-dispersive x-ray spectroscopy (EDS) elemental mapping were carried out on a JEOL JEM-2100 operated at 200 kV. Powder X–ray diffraction (XRD) analysis was operated on Rigaku Rint 2000 X-ray 27 diffractometer with monochromated Cu Kα radiation. Analyses of Zn and Co concentration were measured with inductively coupled plasma optical emission spectrometry (ICP-OES, SPS3520UV-DD, SII nano technology Inc., Japan). Raman spectroscopy acquired with a HORIBA Scientific Lab RAM HR Raman spectrometer system using 532.4-nm laser excitation. Thermogravimetric (TG) analysis was performed on a Hitachi HT-Seiko Instrument Exter 6300 TG/DTA heating from room temperature to 900 °C (5 °C min^{-1}) in nitrogen. The nitrogen adsorption-desorption measurements of the samples were conducted on a Micromeritics BK122T–B analyzer. The specific surface area (SSA) was determined based on Brunauer-Emmett-Teller (BET) theory in the relative pressure range of 0.04 to 0.2. Pore size distributions were determined from the adsorption isotherms, according to the non−local density functional theory (NLDFT) assuming a mixed slit/cylinder pore model. The density was determined by measuring the weight and volume of the active materials.

Electrochemical measurement

Before the electrochemical measurement, the active materials were pressed with 15 Mpa force to decrease the volume. Then, the electrode film was cut into tablets of ~1 cm^2 and pressed onto nickel foam working electrode. The three-electrode system was used to measure the electrochemical performance of the working electrode, with a Pt foil as the counter electrode and a saturated calomel electrode (SCE) as the reference electrode, respectively. All the electrochemical measurements were carried out on a CHI 660D electrochemical workstation. The electrochemical impedance spectroscopy (EIS) measurements were carried out at open circuit potential within the frequency range of 10^{-2} to 10^5 Hz at an AC amplitude of 5 mV. The cycle life tests were conducted by cyclic voltammetry (CV) method at a scan rate of 10 mV s^{-1}. The gravimetric capacitance against the electrode was calculated from the galvanostatic discharge curve based on the following formula:

$$C_{\mathrm{M}} = \frac{I \cdot \Delta t}{m \cdot \Delta V}$$

where I is current, Δt is discharge time, m is the total mass of active material, and ΔV is the voltage variation.

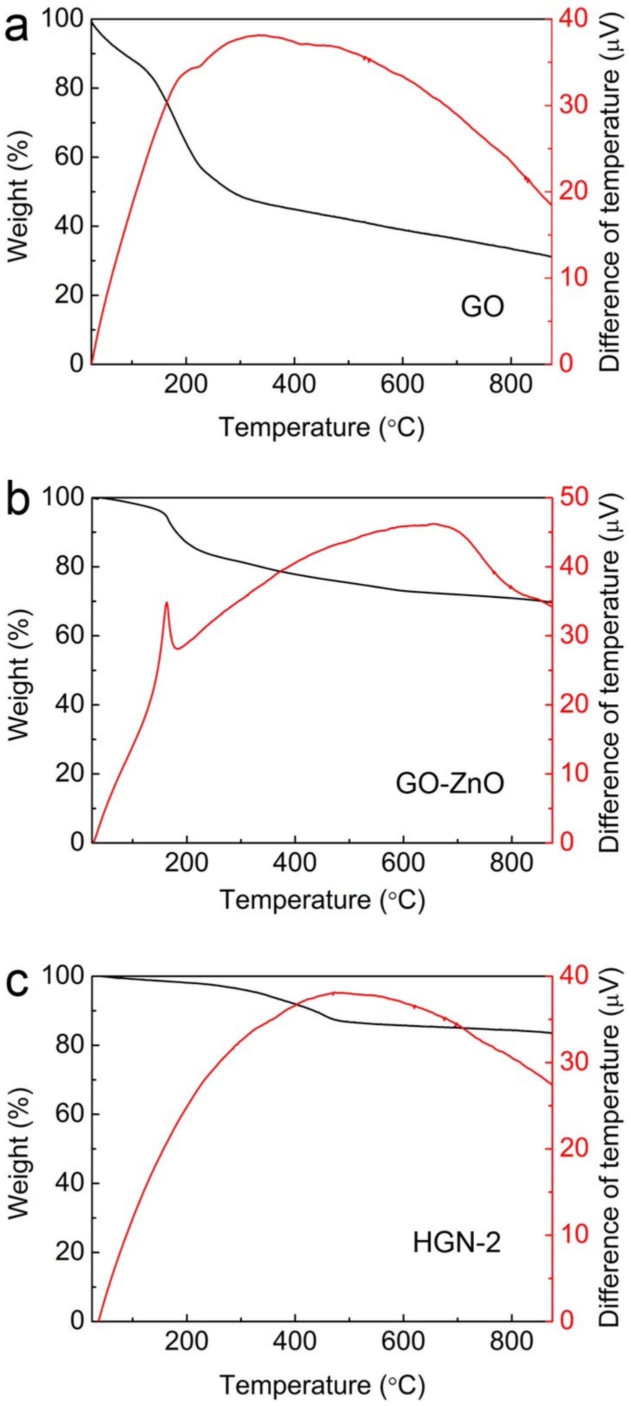

Figure S1. TG-DTA curves of (a) GO, (b) GO/ZnO, and (c) HGN-2 under a nitrogen atmosphere.

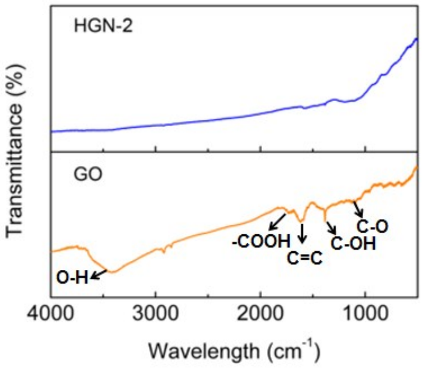

Figure S2. FT-IR spectra of GO and HGN-2.

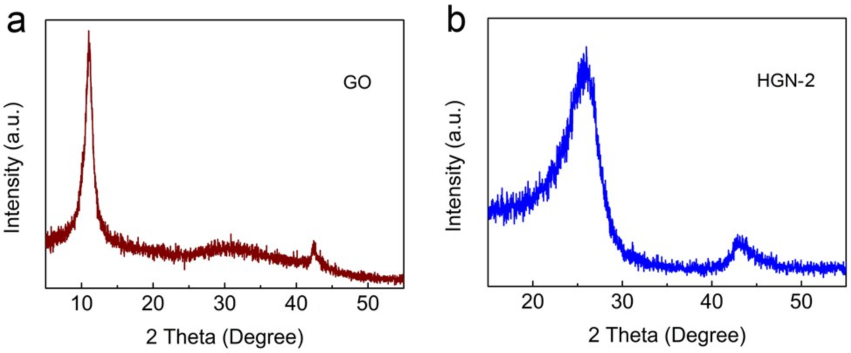

Figure S3. XRD patterns of (a) GO and (b) HGN-2.

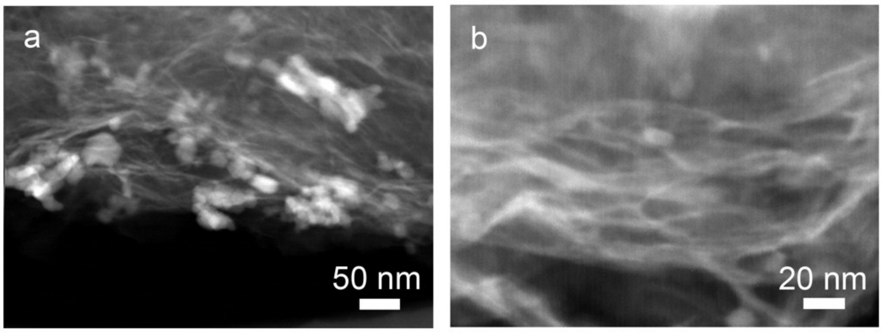

Figure S4. STEM images of (a) GO/ZnO and (b) HGN-2 samples.

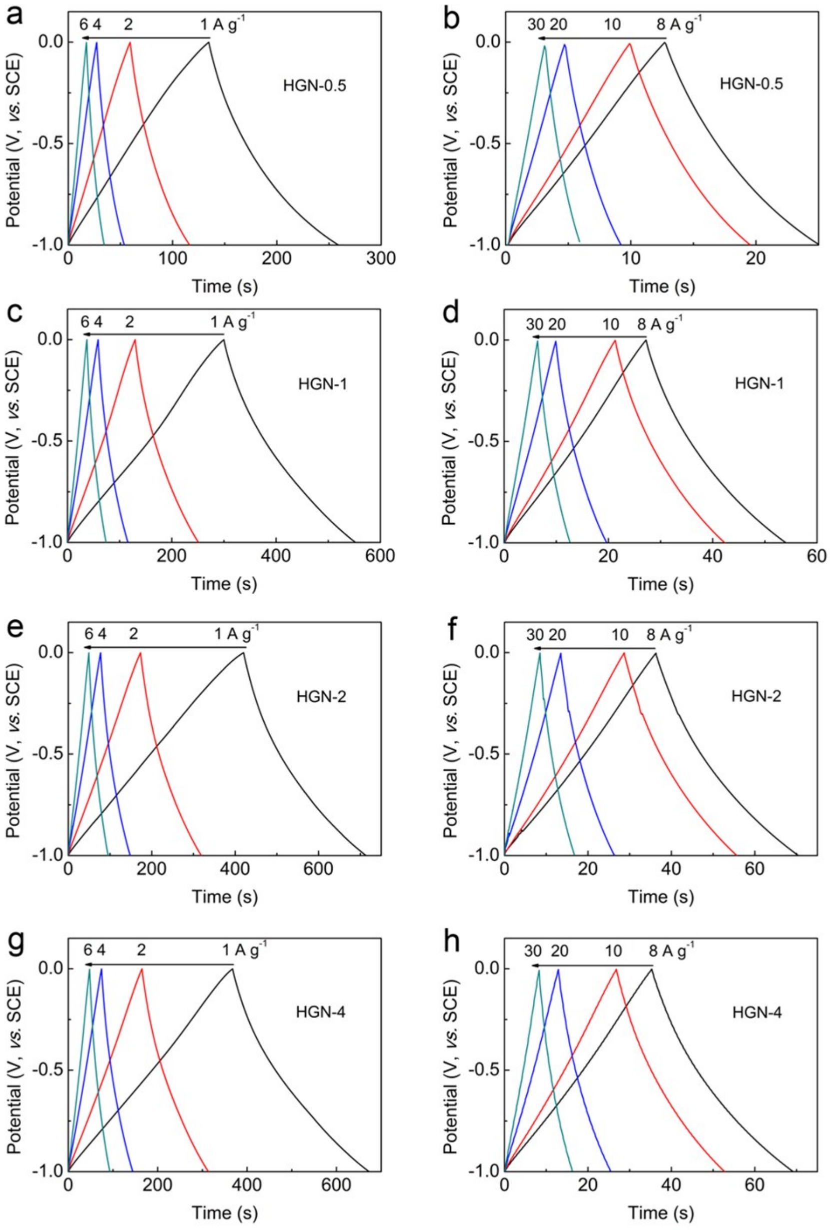

Figure S5. Galvanic charge-discharge (GCD) curves of (a-b) HGN-0.5, (c-d) HGN-1, (e-f) HGN-2, and (g-h) HGN-4.

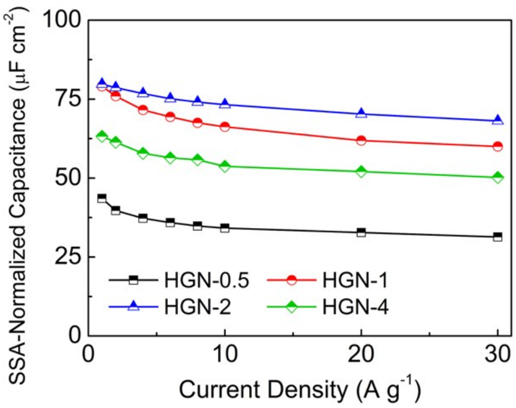

Figure S6. Comparison of SSA-normalized capacitance *vs*. current density.

Table S1. Summary of the surface area (SSA) and pore volume (PV) of HGN-0.5, HGN-1, HGN-2, and HGN-4 samples.

Sample	BET SSA/ $m^2\ g^{-1}$	Micro-SSA/ $m^2\ g^{-1}$	Ratio of Micro-SSA/ BET SSA	Total PV/ $cm^3\ g^{-1}$	Micro-PV/ $cm^3\ g^{-1}$	Ratio of Micro-PV/ Total PV
HGN-0.5	287	83	29%	0.18	0.05	28%
HGN-1	320	113	35%	0.22	0.08	36%
HGN-2	370	207	56%	0.33	0.17	52%
HGN-4	484	329	68%	0.98	0.63	64%

Table S2. Comparison of the C_M and C_V of the HGN-0.5, HGN-1, HGN-2, and HGN-4 electrodes with previously reported graphene-based electrodes and carbon nanosheets electrodes.

Sample name	**Scan rate/ Current density**	**C_M/ F g^{-1}**	**C_V/ F cm^{-3}**	**Electrolyte**	**Ref**
Hydrazine reduced GO	10 mV s^{-1}	123	~143	6M KOH	S2
Reduced GO with high density	1 A g^{-1}	182	255	6M KOH	S3
Porous graphene nanosheets	2 mV s^{-1}	241	101	6M KOH	S4
Holey graphene framework	1 A g^{-1}	310	~220	6M KOH	S5
High density graphene macroform	0.1 A g^{-1}	238	376	6M KOH	S6
Chemically converted graphene	0.1 A g^{-1}	191.7	255	1M H_2SO_4	S7
Porous carbon/graphene composite	1 A g^{-1}	481	212	6M KOH	S8
Functionalized graphene sheets	0.5 A g^{-1}	456	470	6M KOH	S9
Thermal reduced GO	2 mV s^{-1}	276	314	6M KOH	S10
Nitrogen doped carbon nanosheets	2 mV s^{-1}	305	287	6M KOH	S11
HGN-0.5	1 A g^{-1}	125	197	6M KOH	This work
HGN-1	1 A g^{-1}	253	378	6M KOH	
HGN-2	1 A g^{-1}	295	384	6M KOH	
HGN-4	1 A g^{-1}	306	214	6M KOH	

Reference:

1. W. S. Hummers Jr and R. E. Offeman, *J. Am. Chem. Soc.*, 1958, **80**, 1339-1339.
2. J. Yan, T. Wei, B. Shao, F. Ma, Z. Fan, M. Zhang, C. Zheng, Y. Shang, W. Qian and F. Wei, *Carbon*, 2010, **48**, 1731-1737.
3. Y. Li and D. Zhao, *Chem. Commun.*, 2015, **51**, 5598-5601.
4. Z. Fan, Q. Zhao, T. Li, J. Yan, Y. Ren, J. Feng and T. Wei, *Carbon*, 2012, **50**, 1699-1703.
5. Y. Xu, Z. Lin, X. Zhong, X. Huang, N. O. Weiss, Y. Huang and X. Duan, *Nat. Commun.*, 2014, **5**, 4554.
6. Y. Tao, X. Xie, W. Lv, D.-M. Tang, D. Kong, Z. Huang, H. Nishihara, T. Ishii, B. Li and D. Golberg, *Sci. Rep.*, 2013, **3**, 2975.
7. X. Yang, C. Cheng, Y. Wang, L. Qiu and D. Li, *Science*, 2013, **341**, 534-537.
8. J. Yan, Q. Wang, C. Lin, T. Wei and Z. Fan, *Adv. Energy Mater.*, 2014, **4**, 1400500.
9. J. Yan, Q. Wang, T. Wei, L. Jiang, M. Zhang, X. Jing and Z. Fan, *ACS Nano*, 2014, **8**, 4720-4729.
10. L. Jiang, L. Sheng, C. Long, T. Wei and Z. Fan, *Adv. Energy Mater.*, 2015, **5**, 1500771.
11. Q. Wang, J. Yan and Z. Fan, *Electrochim. Acta*, 2014, **146**, 548-555.