Supporting Information

An Evaporation-induced Tri-consistent Assembly Approach towards Ordered Mesoporous Carbon/Graphene Aerogel for High-performance Supercapacitors

Ruili Liu,* Lixia Pan, Xiaoxue Liu and Dongqing Wu*

a. Department of Chemical Engineering, School of Environment and Chemical Engineering, Shanghai University, Shanghai200444, China. E-mail: ruililiu@shu.edu.cn

b. School of Chemistry and Chemical Engineering, Shanghai Jiao Tong University , Shanghai 200240, China. E-mail: wudongqing@sjtu.edu.cn

Experimental Section

Synthesis of GO: GO was prepared from natural graphite powders using a modified Hummers method. Briefly, 3.75 g $NaNO_3$ was firstly mixed with 5 g graphite powders in the flask with a slow addition concentrated 150 ml H_2SO_4. After that, 20 g $KMnO_4$ was gradually added to the mixture with stirring. The mixture were then stand still 5 days and put 500 ml distilled water. Finally, 30 ml H_2O_2 was added until the color of mixture changed to brilliant yellow. The mixture was centrifuged, washed and dialyzed over 7 days with distilled water to remove metal ions and acid. Suspended GO sheets were obtained after ultrasonic treatment.

Synthesis of GA: GAs were synthesized by a combined hydrothermal and freeze-drying process. Typically, a 100 mL GO aqueous dispersion (1.5 mg mL^{-1}) was sealed in a Teflon-lined autoclave and hydrothermally treated at 180 °C for 12 h. Finally, the resulting sample was freeze-dried overnight to obtain GA.

Synthesis of resol precursors: In a typical procedure, 0.61 g of phenol was melted at 42 ^oC and mixed with 0.13 g of 20 wt % NaOH aqueous solution under stirring. After 10 min, 1.05 g of formalin (37 wt %) was added drop wise below 50 ^oC. Upon further stirring for 1 h at 70 ^oC, the mixture was cooled to room temperature and the pH value was adjusted to about 7.0 by 0.2 M HCl solution. After water was removed by vacuum evaporation below 50 ℃, the final product was dissolved in

ethanol (20 wt %).

Synthesis of OMC/GA: 1.6 g Pluronic F127 was dissolved in 8.0 g ethanol with 1.0 g 0.2 M HCl and stirred for 1 h at 40 ℃ to form a clear solution. Then, 5.0 g 20 wt % resols in ethanol solution were added and stirred for 10 min. Next, 2.08 g TEOS was added. After stirring for 2 h, the resultant solution was mixed with GA in a petri dish for 8 h at room temperature to evaporate the ethanol and then for 24 h at 100 ^{o}C for the thermal polymerization of resol. The as-made products were subsequently scraped from the dishes and thermally treated in a tubular furnace at 900 ^{o}C for 2 h under N_2 flow to get mesoporous carbon-silica nanocomposites. After carbon-silica nanocomposites were immersed in 10 wt % HF solutions for 24 h, the composite of mesoporous carbon and GA was obtained and named as GA-OMC.

Electrochemical measurement: Electrochemical measurements were carried out using a CHI 660E electrochemical workstation under ambient condition. In a three-electrode system, 6 M KOH was used as aqueous electrolyte, a platinum plate as the counter electrode, and a Hg/HgO electrode as the reference electrode. To prepare the working electrode, a homogeneous slurry containing polytetrafluoethylene (PTFE) and active materials (weight ratio 5:95, in ethanol) was painted on the nickel foam and pressed under 15 MPa face to face for 5 minutes. The electrode was then dried at 100 ^{o}C in vacuum overnight.

Characterization: The obtained products were characterized by the small-angle X-ray scattering (SAXS, Cu KR radiation, 40 kV, 35 mA)*;* x-ray diffraction (XRD, 3KW D/MAX2200V PC, Cu Ka radiation), field-emission scanning electron microscopy (FE-SEM, JSM-6700F), transmission electron microscopy (TEM, JEOL JEM-2010F), and Raman spectra (INVIA) in the Instrumental Analysis and Research Center, Shanghai University. Using the DFT methods, the pore volumes and pore size distributions were derived from the adsorption branches of isotherms. The Brunauer-Emmet- Teller(BET) method was utilized to calculate the specific surface area.

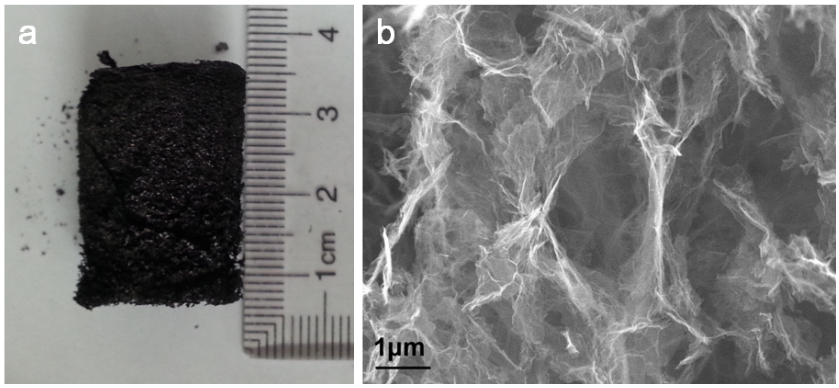

Figure S1. (a) Optical image of OMC/GA and (b) SEM image of GA

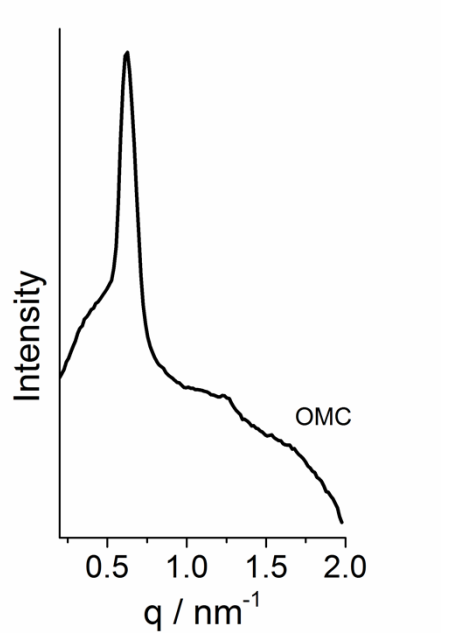

Figure S2. SAXS pattern of OMC

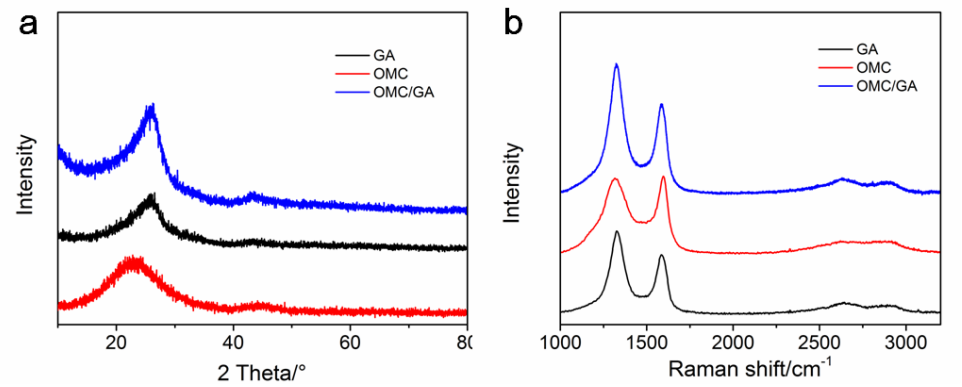

Figure S3. (a) XRD patterns and (b) Raman spectra of OMC/GA, GA and OMC

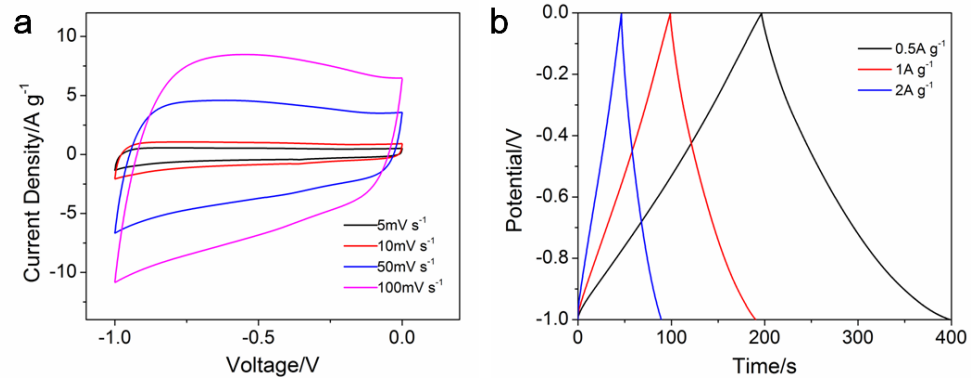

Figure S4. (a) CVs and (b) galvanostatic charging/discharging curves of the GA.

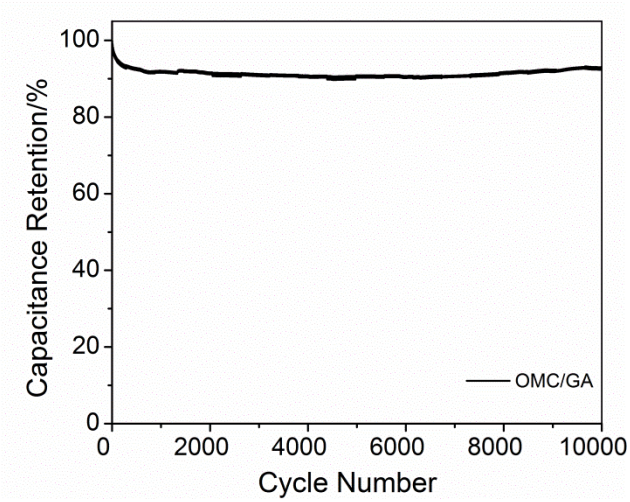

Figure S5. Cycling stability of OMC/GA electrodes.

Table S1. Porosity and electrochemical properties of OMC, GA and OMC/GA

	Pore volume ($cm^3\ g^{-1}$)	**Surface area** ($m^2\ g^{-1}$)	**Capacitance** (F g^{-1} at 100 mV s^{-1})	**Durability after 1000 cycles**
OMC	2.08	2470	102	80%
GA	0.45	220	67	93%
OMC/GA	0.99	715	115	94%