Supporting Information for

Graphene-based mesoporous nanocomposites of spherical shape with 2-dimensional layered structure

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Experimental

In this paper, graphene oxide—which was synthesized using the modified Hummer's method, and expanded graphite (Asbury carbon)—was used as the starting material in the synthesis of the mesoporous M-RGO/SiO_2 composites.

1. Synthesis of M-RGO/SiO_2

The mother liquor including these starting materials was prepared as follows: One gram (for M-RGO/SiO_2) or 5 g (for M-RGO/SiO_2-H) of a tri-block copolymer ($(EO)_{20}$-$(PO)_{70}$-$(EO)_{20}$, Aldrich) was dissolved as a pore-structure agent in an acidic solvent containing nitric acid (5 g), ethanol (30 ml), and distilled water (50 ml) (solution 1). The acidic conditions in this solvent led to high levels of hydrolysis, and few condensation reactions. Graphene oxide (0.05 g) was dispersed in distilled water (20 ml), using bath sonication for 30 min (solution 2). Solution 2 was then dropped into solution 1. Finally, 0.2 M of tetraethylorthosilicate (TEOS, Aldrich, 98 %) was dissolved in this mixed solution. After stirring the solution for 1 hr, the M-RGO/SiO_2 samples were synthesized in the ultrasonic spray reactor, which had a temperature of 773 K; nitrogen (1 L/min) was used as the carrier gas. The details of the ultrasonic spray reactor have been reported elsewhere.[1] The prepared samples were calcined for 2 hr, using a tube furnace at 773 K, in nitrogen gas with a flow of 2.5 L/min.

2. Synthesis of M-RGO/carbon

The M-RGO/Carbon and M-RGO/Carbon-H were prepared using a two-step template-synthesis technique: the M-RGO/SiO_2 (template for M-RGO/carbon) and M-RGO/SiO_2-H (template for M-RGO/Carbon-H) prepared using spray pyrolysis were impregnated in an aqueous solution made from distilled water of five times the template weight and along with 80 % sucrose and 10 % sulfuric acid (DC Chemical Co., Ltd., 98 % solution) as much as the pore volume of the template. The mixture was stirred and then dried in an oven for 3 h at 373 K, and subsequently at 413 K for another 6 h. The resulting black powder was treated again using the above-mentioned step; with 40% sucrose and 10% sulfuric acid based on the pore volume of the template and distilled water of five times the template weight. The obtained powders were heated consecutively at 723 K and 973 K, for 3 h and 2 h, respectively, to carbonize the carbon source. After pyrolysis, the SiO_2-carbon composite was treated twice with a 0.2 M solution of NaOH in a 50/50 mixture of water and ethanol, to remove the SiO_2.[2]

3. Characterization

The morphology and microstructure of the samples were observed using field emission transmission electron microscopy (FE-TEM, Tecnai G2 F30, 300 kV), and field emission SEM (Magellan 400). The atomic distribution was determined using an electron energy loss spectrometry (EELS) mapping method in HR-TEM (HR-[S] TEM-2200FS with image Cs-corrector). To gain knowledge about the chemical bonding in the samples, XPS (Sigma Probe) analysis was used. The specific surface area was determined using the Brunauer-Emmett-Teller (BET) method, while the pore sizes and volumes were calculated using the Barrett-Joyner-Halenda (BJH) method (Supporting Data).

The specific capacitances were recycled 20 times and carried out using a typical three-electrode experimental cell equipped with a working electrode, a platinum foil counter electrode and a standard calomel electrode (SCE), reference electrode, in 1M H_2SO_4 solution as electrolyte on Cyclic voltammetry (Solartron 1480). The working electrode materials composed of synthesized samples and binder (PVDF) with a weight ratio of 8:2 were coated on the titanium substrate. It didn’t contain any carbon black as the conducting material. The surface resistance of paste as working electrode materials was coated on PET film and its resulting value was about 4×10^7 Ω/□.

Supplementary References

(1) Song, L.; Park, S. B. *J. Nanosci. Nanotechnol*. **2010**, *10*, 122.

(2) Jun, S.; Joo, S. H.; Ryoo, R.; Kruk, M.; Jaroniec, M.; Liu, Z.; Ohsuna, T.; Terasaki, O. *J. Am. Chem. Soc*. **2000**, *122*, 10712.

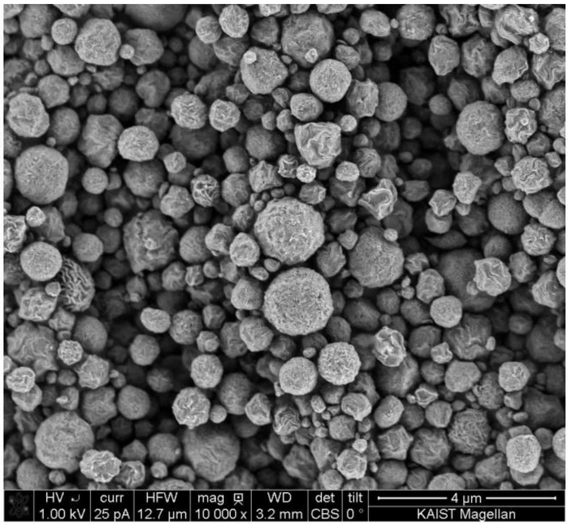

Figure S1.Overall morphology of Sample (M-RGO/SiO_2-H) by SEM at lower magnification.

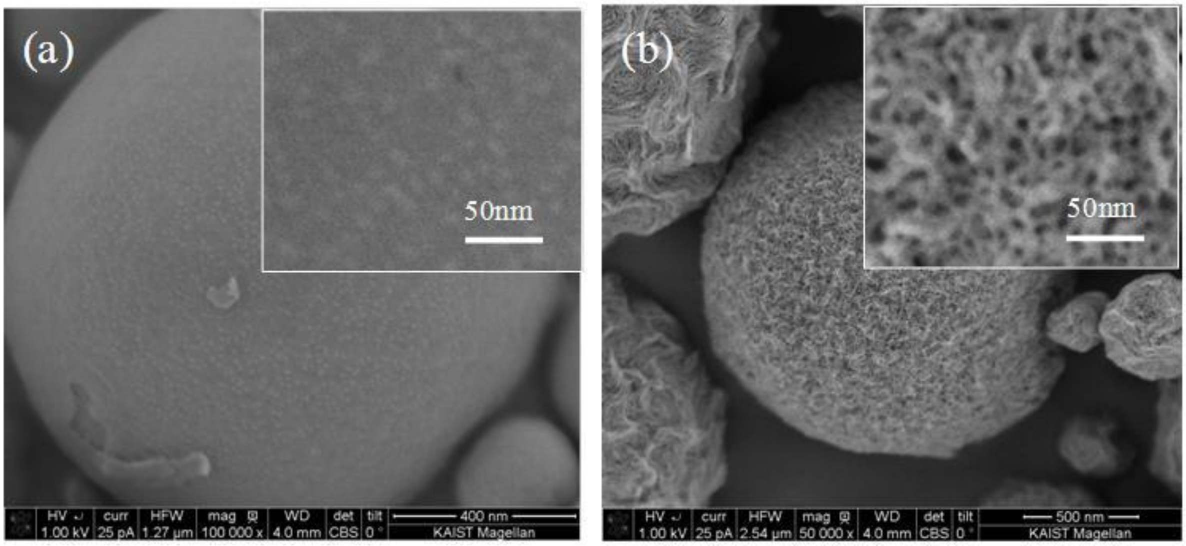

Figure S2. Surface images of (a) M-SiO_2 without RGO and (b) M-RGO/SiO_2 using a high concentration of pore-agents (M-RGO/SiO_2-H)

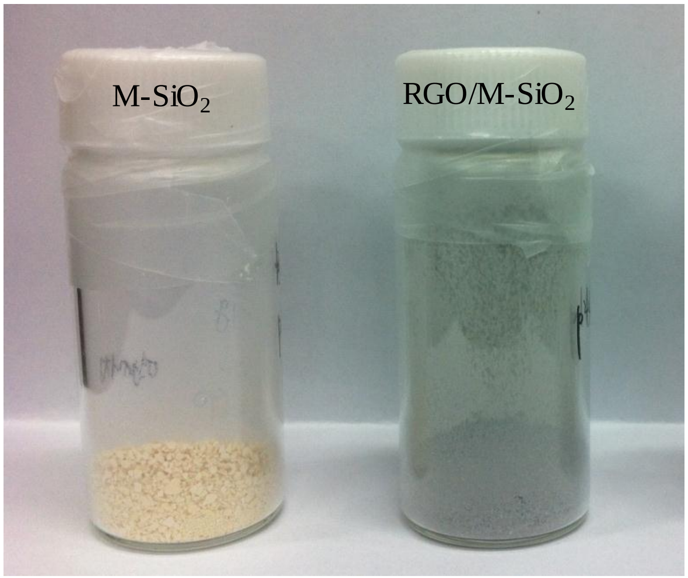

Figure S3. Photographs of M-SiO$_2$ without RGO and M-RGO/SiO$_2$

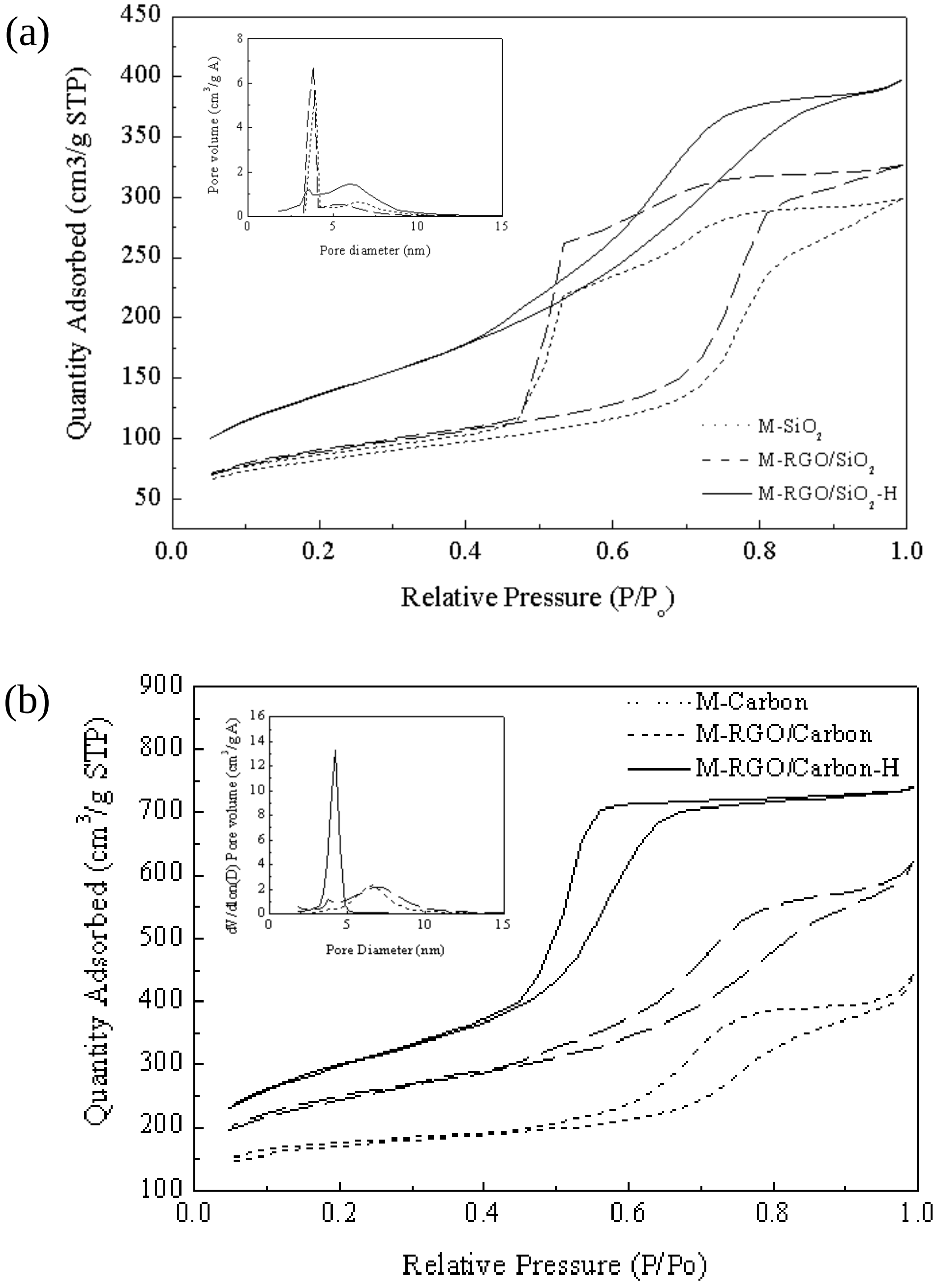

Figure S4. Nitrogen adsorption isotherms and pore size distributions (insert) of samples

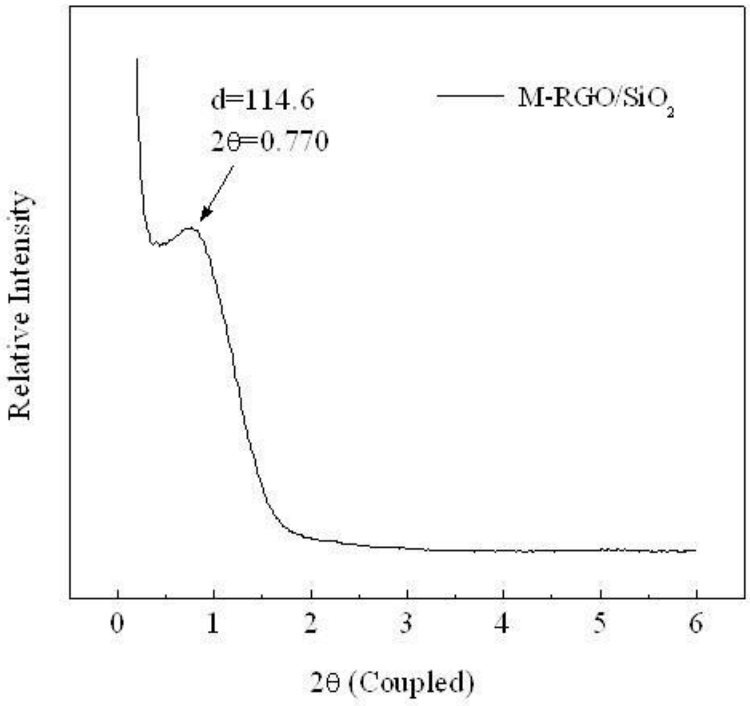

Figure S5. Small angle XRD data of M-RGO/SiO_2

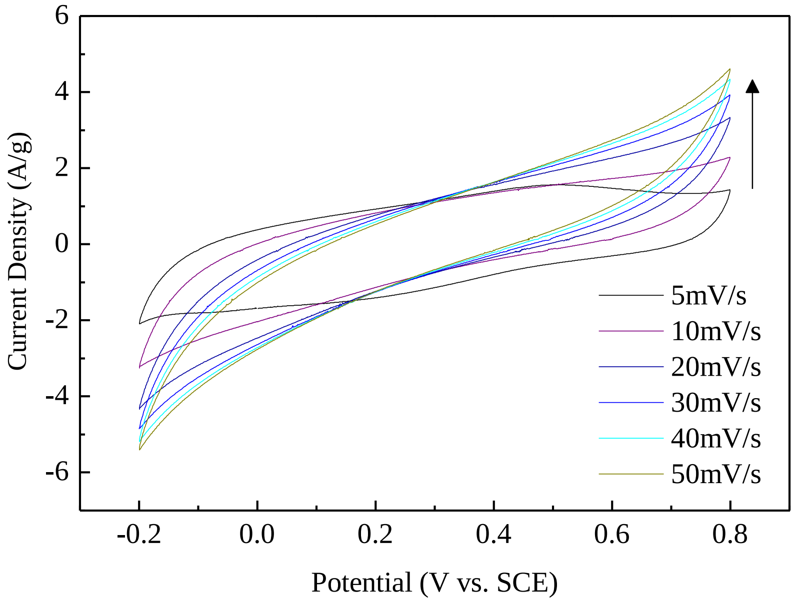

Figure S6. Cyclic voltammetric profiles of M-RGO/Carbon-H at various scan rate

The shape of the CV loop at scan rate of 5mV/s is close to a rectangle. However, the cure shape is gradually distorted from the rectangular with increasing the scan rates as shown Figure S6. It means that the internal resistance of working electrode is increased as the scan rates increase. In this paper, the working electrode was only composed of M-RGO/Carbon and binder without any conducting materials. In addition, the surface resistance (≒ 10^7 Ω/□) was affected by the mesoporous carbon on the graphene. Therefore, further study on the optimum process of the formulation of paste for electrode as well as its carbonization and reduction is needed. These questions will be addressed in future work.

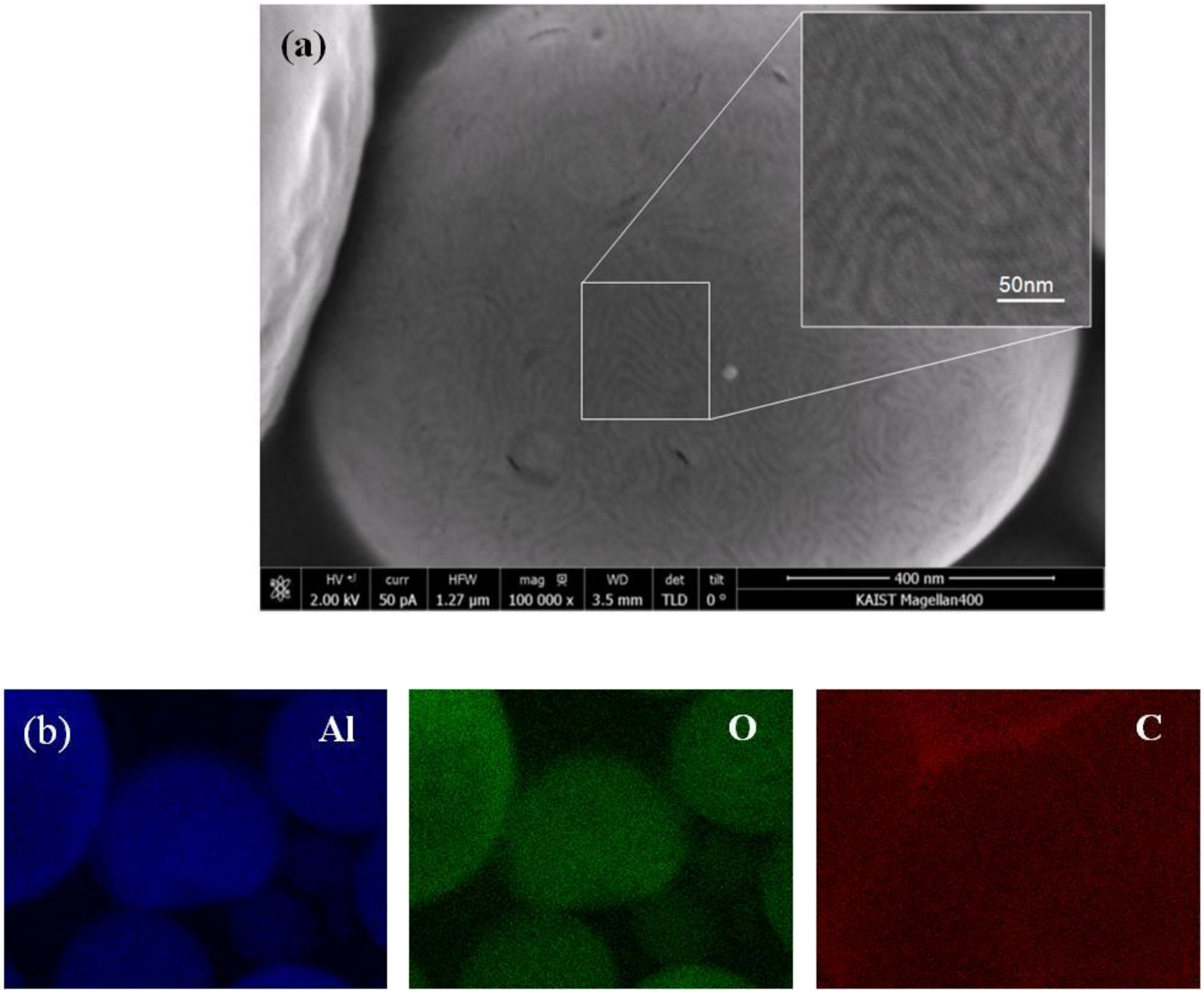

Figure S7. (a) Surface image and (b) EDS mapping images of M-alumina/RGO particles by SEM

Table S1. Surface area, pore size and pore volume of samples

	BET (m^2/g)	Pore size[1] (nm)	Pore volume[2] (cm^3/g)
M-SiO_2	295	4.5	0.47
M-RGO/SiO_2	316	4.1	0.51
M-RGO/SiO_2 -H	494	4.5	0.61
M-Carbon	627	6.5	0.58
M-RGO/Carbon	879	5.2	0.90
M-RGO/Carbon -H	1057	4.1	1.17

[1] BJH Desorption average pore diameter (4V/A)

[2] BJH Desorption cumulative volume of pores between 1.7nm and 300 nm diameter