Electronic Supplementary Information (ESI) for:

Gram-scale Synthesis of Nanomesh Graphene with High Surface Area and Its Applications in Capacitor Electrodes

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1. Experimental methods

1.1 Preparation of MgO template

First, purchased MgO powder (Sinopharm Chemical Reagent Co. Ltd.) was mixed with deionized water accompanied with ultrasonic agitation. The mixture was boiled for 2 to 24 hr in a reflux apparatus. After filtration and drying, the material obtained was ground into a fine powder. Finally, the powder was calcined at 500°C for 30 min to remove water.
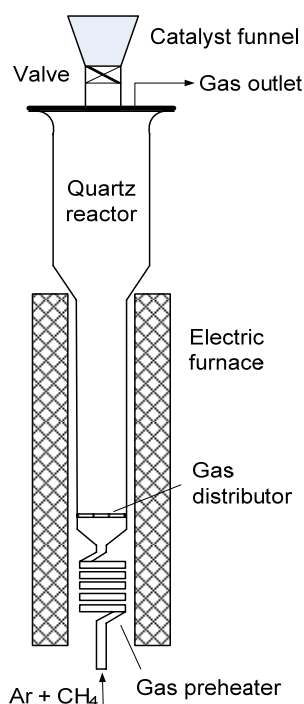
1.2 Synthesis process of nanomesh graphene

Fig. S1 Schematic of the downer reactor for the synthesis of the porous graphene.

Porous graphene was synthesized by CH_4 cracking at 900 °C in a vertical quartz reactor with diameter of 50 mm and length of 1500 mm shown in Fig. S1. A sintered porous plate was used as the gas distributor at the bottom of the reactor. Gas entered the bottom preheater of the reactor, then passed through the gas distributor, and finally flowed out into the atmosphere. On the top of the quartz reactor, there was a stainless steel plate with a hopper for feeding catalyst. In a typical run, the quartz reactor was mounted in an electrical tube furnace and was heated to 900°C in an argon flow of 1000 ml/min at atmospheric pressure. When the reaction temperature was reached,

800 ml/min CH_4 was introduced into the reactor. Then, the MgO template (about 30 g) was fed into the reactor over 5 min from the top hopper. After 10 min carbon deposition, CH_4 was turned off and the reactor was cooled to room temperature in an Ar atmosphere. The material obtained was purified by acid washing (excessive amount of condensed hydrochloric acid diluted by deionized water, with volume ratio of 1:1) with reflux for 1 hour to remove MgO. This was followed by filtration and drying at 80 °C overnight in an oven. The material obtained was ground into a fine powder for characterization and application.

1.3 Electrochemical testing

Electrodes for supercapacitors were prepared by mixing the porous graphene, carbon black and poly(tetrafluoroethylene) in a mass ratio of 85:10:5 to give a slurry. The slurry was pressed onto a nickel foam current collector (1 cm^2) and dried at 100°C for 12 h. All electrochemical measurements were made in a three-electrode setup, in which platinum foil and Hg/HgO electrodes (saturated calomel electrode, SCE) were used as counter and reference electrodes. Cyclic voltammetry (CV) and galvanostatic charge/discharge were measured by a CHI 660C electrochemical workstation in 6 M KOH electrolyte. The specific capacitance of the electrode was calculated by $C = (\int IdV)/(\upsilon mV)$, where I was the response current density (A), V was the potential (V), υ was the potential scan rate (mV s^{-1}), and m was the mass of the electroactive materials in the electrodes (g).

1.4 Characterization

The samples for transmission electron microscope (TEM), atomic force microscope (AFM) and scanning transmission electron microscopy (STEM) characterization were prepared by a small amount of purified graphene dispersed in ethanol by sonication for 10 min to form a homogeneous suspension. A drop of the suspension was deposited on a carbon-coated copper grid for TEM observation (JEM 2010 microscope, operated at 120.0 kV) to get a general view of the material. Centrifugation was used to remove large pieces of the material from the supernatant. A drop of the supernatant was deposited on a mica substrate for AFM observation (SPM-9600), and on a carbon-coated copper grid for STEM observation (FEI Tecnai G^2 F20, operated at 200 kV).
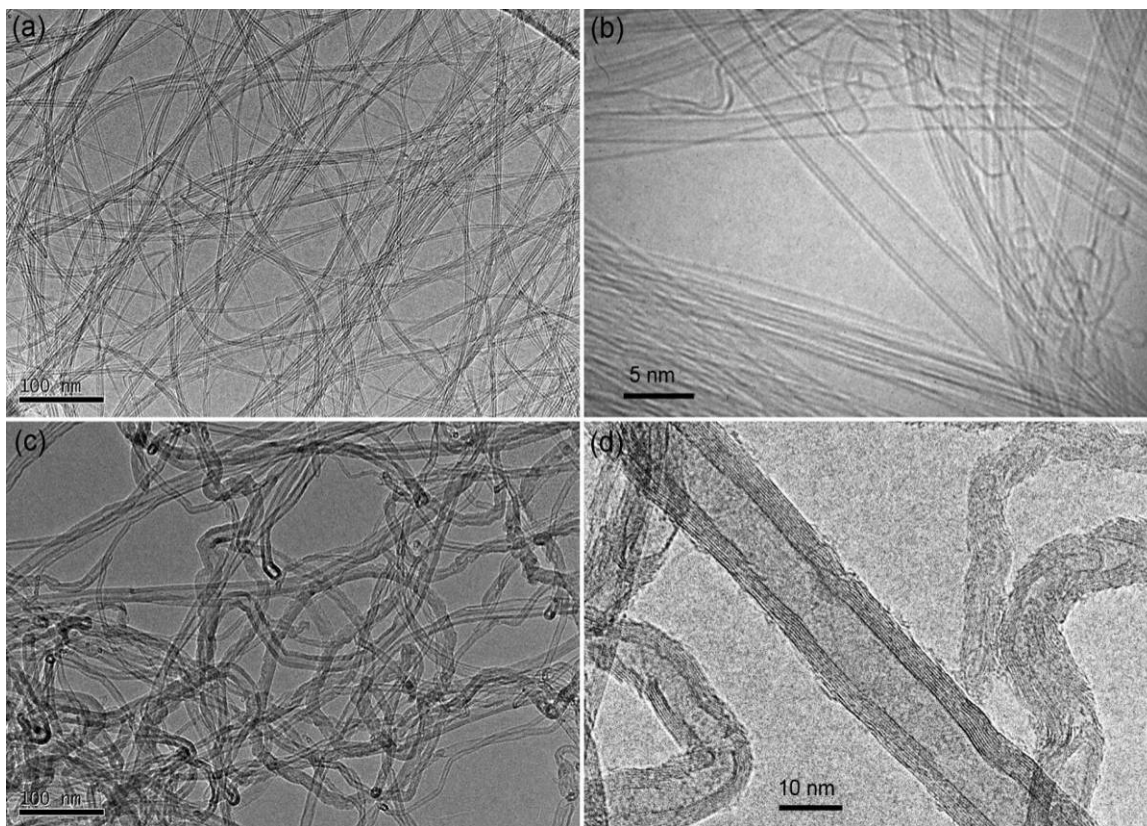
Fig. S2 TEM photos of (a, b) double walled carbon nanotubes (DWCNTs) and (c, d) multi-walled carbon nanotubes (MWCNTs) used for Raman calibration.

Raman measurements at 633 nm excitation were performed with a Renishaw RM2000 Raman spectrophotometer. DWCNTs, MWCNTs and natural graphite were used for Raman calibration. The DWCNTs were synthesized by methane cracking on Fe/MgO catalysts at 900°C.[1] In the DWCNT sample, there was 70% DWCNTs, with the remainder being SWCNTs (Fig. S2a and b). Neither MWCNTs nor carbon nanofibers (CNFs) were found in wide view field TEM observations. The DWCNT sample had a diameter distribution from 1 to 5 nm with an average of 2-3 nm. The MWCNTs were prepared by ethylene cracking in a nano-agglomerate fluidized bed reactor.[2] The MWCNTs had a diameter distribution from 5 to 20 nm with an average of 10.5 nm. The layer numbers of the MWCNTs were in the range of 5-10 (Fig. S2c and d). The natural graphite was a purchased chemical with more than 10 graphene layers.

Thermogravimetric analysis (TGA) was performed on a Q500 in air flow with a temperature scan of 10°C/min. X-ray diffraction (XRD) data were obtained on a D8 ADVANCE diffractometer utilizing the Cu Kα1 line ($\lambda = 0.15406$ nm). The pore size distribution and specific

surface area (SSA) were measured on a Micrometritics ASAP 2010 using nitrogen as adsorptive. A degasification at 300°C (or 600-100°C for calcination) for 10 min was performed before the nitrogen adsorption. The mesopore distribution (1.7-300 nm) was calculated by the Barrett-Joyner-Halenda (BJH) method. The micropore distribution (0.5-1.8 nm) was calculated by the original Horvath-Kawazoe (HK) method. The SSA was calculated by the Brunauer-Emmett-Teller (BET) method. X-ray photoelectron spectroscopy (XPS) measurements were performed on a PHI Quantera Scanning X-ray Microprobe.

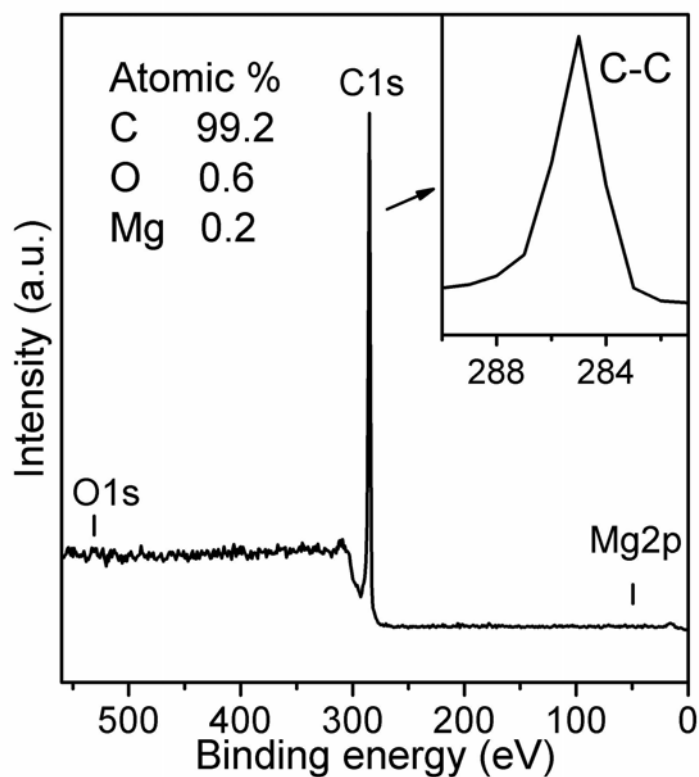## 2. Characterization data of nanomesh graphene

Fig. S3 XPS data of the nanomesh graphene. More than 99 atomic% on the surface of the purified nanomesh graphene is carbon, and no graphite oxide exists.
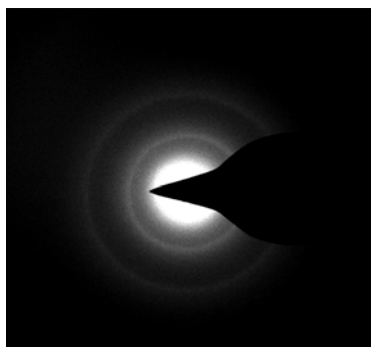

Fig. S4 A typical selected area electron diffraction (SAED) pattern of the nanomesh graphene. The rings corresponding to a multicrystal show the crystalline nature of the sample.
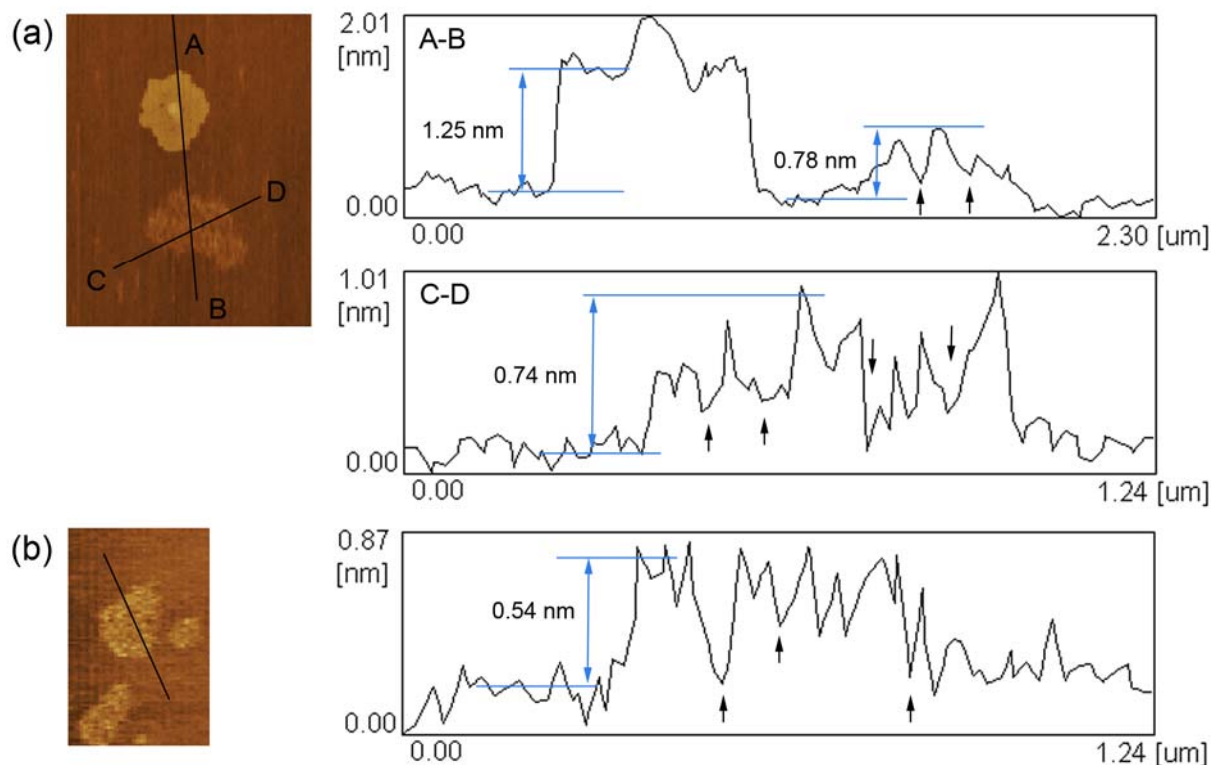
Fig. S5 AFM measurement of the nanomesh graphene sheets. The black arrows indicate the existence of the mesh structure. The graphene sheets deposited on the two surfaces of one MgO layer were piled together as one combined graphene sheet with a height of ~1.25 nm, as shown in the top part of a. Single graphene layers with a height of 0.5-0.8 nm were present in a (bottom) and b. The central raised part of the top graphene sheet in (a) was inferred to be MgO or $MgCl_2$ residues that remained between the two single graphene sheets.
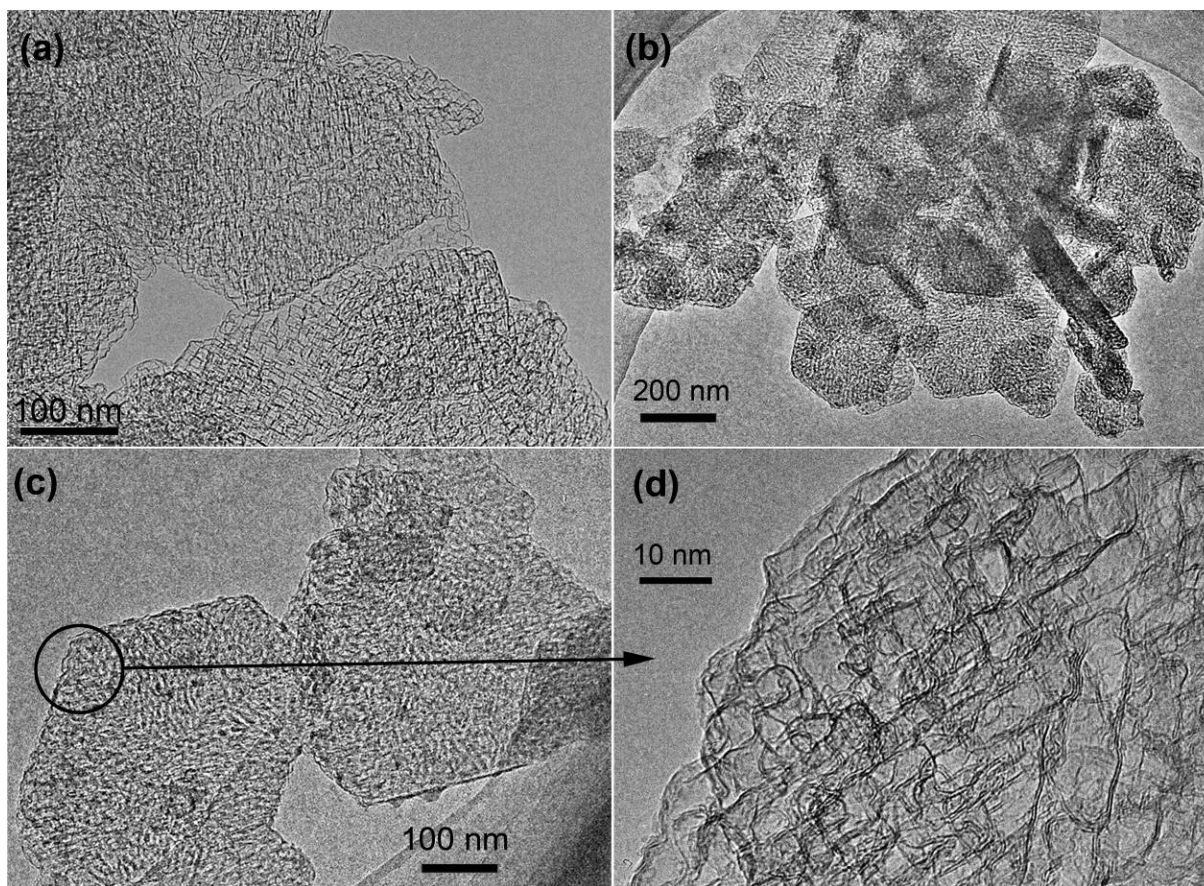
Fig. S6 TEM photos showing the staking morphologies of the nanomesh graphene sheets. (a) Remarkable corrugations in the porous graphene sheets; (b) the stacking manner of the graphene. (c) The graphene sheets deposited on the two surfaces of one MgO layer were piled together as one combined graphene sheet. In a closeup view of the edge of a combined graphene sheet (d), slight differences in focus can be found on the two single graphene sheets, and no dark multi-lines of graphene with more than two layers were observed. These results indicated that there was a distance much larger than 0.4 nm between the two single graphene sheets, which showed a different appearance from that of multi-layered graphene.

Reference

(1) G. Ning, Y. Liu, F. Wei, Q. WenandG. Luo, *J. Phys. Chem. C*, 2007, **111**, 1969.

(2) Y. Wang, F. Wei, G. Luo, H. YuandG. Gu, *Chem. Phys. Lett.*, 2002, **364**, 568.