Electronic Supplementary Information

One-Step Synthesis of Nitrogen-Doped Graphene-Like Meso-Macroporous Carbons as Highly Efficient and Selective Absorbents for CO_2 Capture

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EXPERIMENTAL DETAILS

Chemicals

Dicyandiamide, melamine and glucose were supplied by Sigma Aldrich Co. Ltd., USA. All reagents were of analytical grade and used as received without further purification.

Materials synthesis

Nitrogen-doped graphene-like meso-macroporous carbons (GMCs) were synthesized by carbonizing the mixture of dicyandiamide/glucose or melamine/glucose. Typically, 4.0 g of melamine and 0.1 g of glucose were mixed well, then the mixture was transferred into the tube furnace under flowing N_2. The temperature of the tube furnace experienced the following heating procedures: from room temperature to 600°C (2.5°C/min), from 600°C to 800°C (1.67°C/min), and remained at 800°C for 1 h. Other GMCs were synthesized by similar procedures.

In the synthetic process, dicyandiamide (or melamine) has two roles: nitrogen source and template. The pyrolysis of dicyandiamide (or melamine) at 600ºC would result in layered carbon nitride (g-C_3N_4), which can bind the as-formed carbon intermediates through donor-acceptor interactions and act as the template to direct the formation of carbon nanosheets. The complete thermolysis of g-C_3N_4 template at the final target temperature (700 or 800ºC) leaves the pure nitrogen-doped GMCs. For the purpose of forming g-C_3N_4 template and removing g-C_3N_4 template, two different heating rates were applied before and after 600ºC. The heating rate of 2ºC/min before 600ºC was selected in accordance with the synthetic process of g-C_3N_4 from dicyandiamide (or melamine) reported in the literature, while the heating rate of 1.67ºC/min after 600ºC was selected to ensure the complete thermolysis of g-C_3N_4 template.

Characterization

The N_2 adsorption isotherms at −196°C were measured on a Micromeritics Gemini 2390t system to determine the textural properties. Before the measurement, the samples were pretreated at 150°C under high vacuum (0.1 mbar) for 12 h. The total surface area was calculated from the N_2 adsorption isotherms using the BET equation in the relative pressure range of 0.05–0.20. The total pore volume was determined from the amount of N_2 adsorbed at the relative pressure of 0.97. The micropore volume was calculated by the *t*-plot method in the thickness range of 0.45–0.6 nm. The pore size distribution was derived from the N_2 adsorption isotherms using the BJH model. Elemental analysis was performed on an Elementar Vario EL II analyzer. X-ray diffraction (XRD) patterns were collected on a PANalytical powder diffractometer using Cu Kα radiation at the voltage of 40 KV and current of 40 mA. Raman spectra were collected on a Princeton MSL 532-50 spectrometoer with λ=532 nm laser excitation. X-ray photoelectron spectroscopy (XPS) spectra were collected on a Thermo ESCALAB250 spectrometer with Al Kα radiation, and the binding energies were calibrated using the C1s peak at 284.9 eV. Scanning electron microscope (SEM) images were taken on a Zeiss Auriga Crossbeam SEM at an acceleration voltage of 5 kV. Transmission electron microscope (TEM) images were

taken on a Tecnai 30 field-induced electron microscope (FEI, Netherlands) with an acceleration voltage of 200 kV.

Gas adsorption measurements

CO_2 and N_2 adsorption isotherms at 0 and 25°C were measured on a Micromeritics Gemini 2390t surface area analyzer. Before the measurement, samples were pretreated at 150°C under flowing N_2 for 12 h.

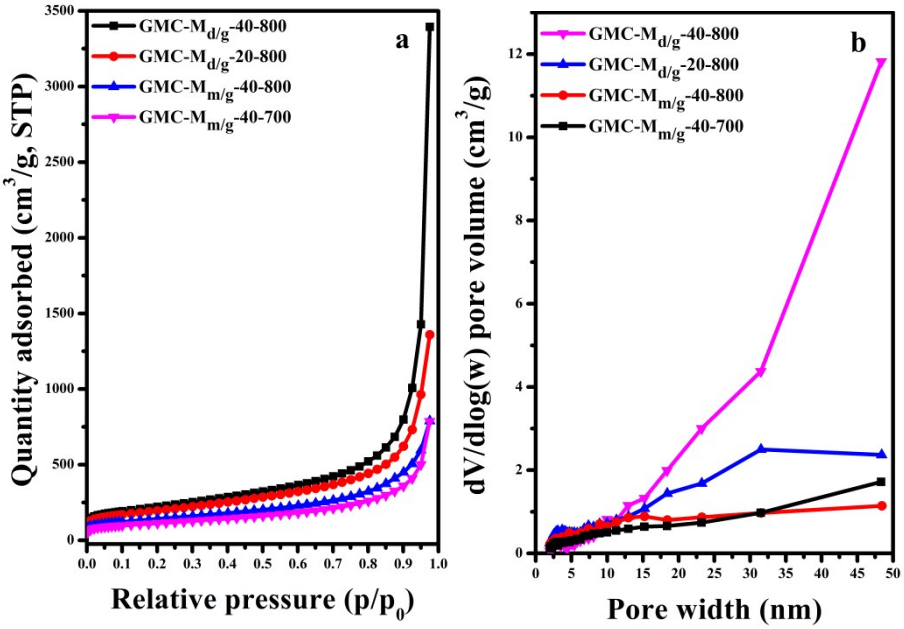

Fig. S1 N_2 adsorption isotherms at -196°C (a) and BJH pore size distributions (b) of synthesized GMCs.

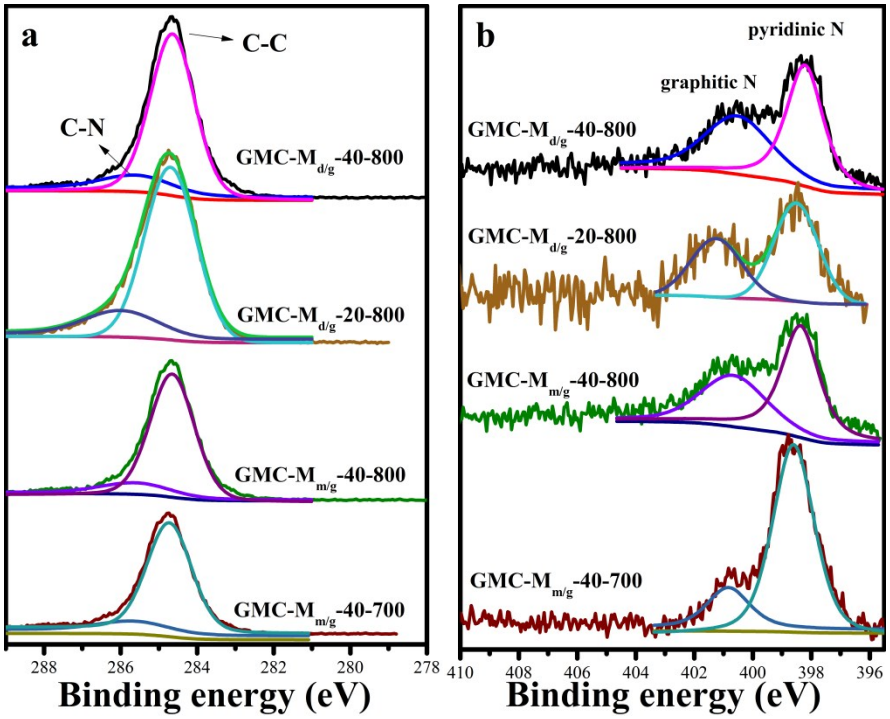

Fig. S2 C 1s (a) and N 1s (b) spectra of synthesized GMCs.

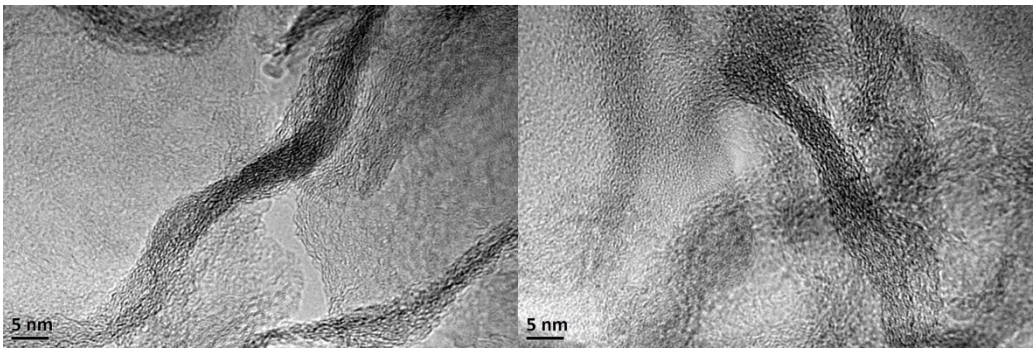

Fig. S3 HR-TEM images of GMC-$M_{d/g}$-40-800.

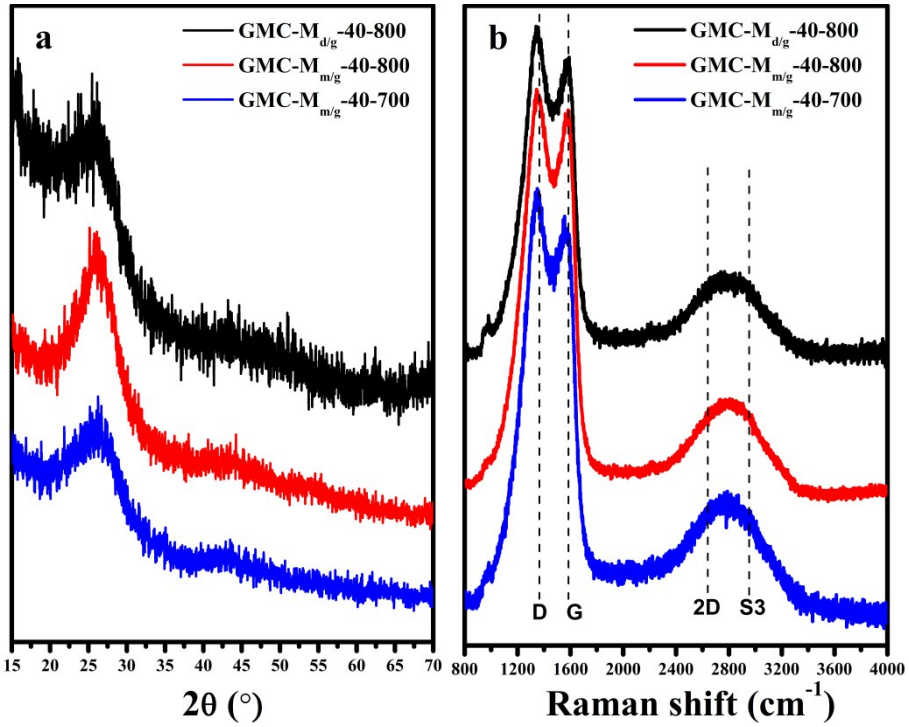

Fig. S4 XRD patterns (a) and Raman spectra (b) of synthesized GMCs.

Table S1. CO_2 capacities of synthesized GMCs at different temperatures and pressures

Samples	CO_2 capacities at 0°C (mmol/g)		CO_2 capacities at 25°C (mmol/g)	
	0.15 bar	1 bar	0.15 bar	1 bar
GMC-$M_{d/g}$-40-800	1.51	3.19	1.49	2.78
GMC-$M_{d/g}$-20-800	1.82	2.95	1.34	2.30
GMC-$M_{m/g}$-40-800	1.08	1.77	0.85	1.43
GMC-$M_{m/g}$-40-700	2.18	3.43	1.63	2.66