

## Supporting Information

# A Facile Solvent-Free Route to Synthesize Ordered Mesoporous Carbons

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## Experimental section

*Synthesis of ordered mesoporous carbon (H-OMC):* H-OMC was synthesized through solid-phase synthesis route as following: 1.0 g of triblock copolymer Pluronic F127 was added in agate mortar ( $\varnothing$  15 cm) at room temperature and ground into fine powder. Then 0.22 g of resorcinol was added and ground in the presence of F127. 0.40 g of terephthalaldehyde was added in 3 batches and the mixture was intensely ground with pestle for 5 minutes during which sticky polymer was formed. The sticky nanocomposite was placed in a quartz boat at 100 °C for 8 hours to obtain as-made product, which was finally carbonized in a tubular furnace at 600 °C for 3 h under nitrogen flow (flow rate of 100 mL/min) at the ramp of 1 °C/min. C-OMC, N-OMC, Mo-OMC and F-OMC were synthesized followed the same way, besides adding different amount kinds of precursors. All of the as-made samples were carbonized under nitrogen flow, unless the preparation of Mo-OMC, which was carbonized under hydrogen flow at 600 °C for 3 h (flow rate of 100 mL/min) at the ramp of 1 °C/min. The details of preparation of all samples were listed in Table S1.

## Characterization

Small angle X-ray diffraction (XRD) patterns were taken on a Bruker D8 X-ray diffractometer with Ni-filtered Cu K $\alpha$  radiation (40 kV, 40 mA). The d-spacing values were calculated using the Bragg's diffraction formula of  $2d\sin\theta$

=  $n\lambda$ , and the unit parameters unit parameters ( $a$ ) were calculated from the formula  $a = 2d_{110}/3^{1/2}$  for OMC and  $a = d_{110} \cdot 2^{1/2}$  for cubic carbon of C-OMC. The N<sub>2</sub> adsorption-desorption isotherms were carried out at 77 K on a Micromeritics TriStar 3000 apparatus at -196 °C. Before analysis, the tested samples were degassed at 473 K for 6 hours under vacuum. The surface area was calculated by the Brunauer-Emmett-Teller (BET) method and the pore size distribution curve was calculated by the Barret-Joyner-Halenda (BJH) method using adsorption branch of the isotherms. And the total pore volumes ( $V_t$ ) were estimated from the adsorbed amount of nitrogen at a relative pressure  $P/P_0$  of 0.995. The micropore volumes ( $V_m$ ) was calculated from the V-t plot method using the equation of  $V_m/\text{cm}^3 = 0.001547I$ , where I represents the y intercepts in the V-t plots. The t values were calculated as a function of the relative pressure using the de Bore equation,  $t/\text{Å} = [13.99/(\log(P_0/P) + 0.0340)]^{1/2}$ . Transmission electron microscopy (TEM) experiments were conducted on a JEOL 2011 microscope (Japan) operated at 200 kV. The samples for TEM tests were suspended in ethanol and supported onto a holey carbon film on a Cu grid.

**Table S1. Preparation conditions of H-OMC, C-OMC, N-OMC, Mo-OMC and F-OMC nanocomposites prepared through solid-phase synthesis method**

Sample	F127/g	Resorcinol /g	Terephthalaldehyde/ g	Melamine /g <sup>a</sup>	Mo source /g <sup>b</sup>
H-OMC	0.62	0.22	0.40	-	-
C-OMC	0.41	0.22	0.40	-	-
N-OMC	1.50	0.44	0.56	2.0	-
Mo-OMC	0.62	0.22	0.40	-	0.030
F-OMC	0.62	0.44	0.49 <sup>c</sup>	-	-

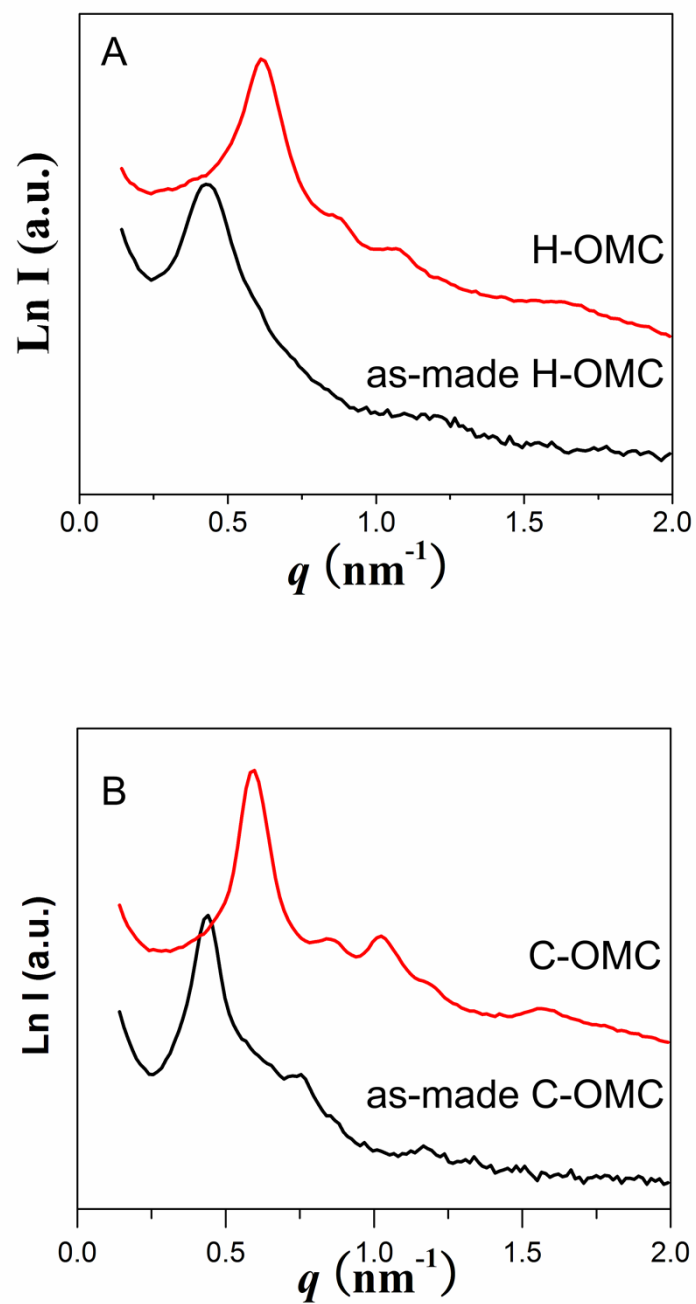
<sup>a</sup>Melamine served as N source.

<sup>b</sup>Phosphomolybdic acid served as Mo source and the nominal content of Mo was ~5 wt%.

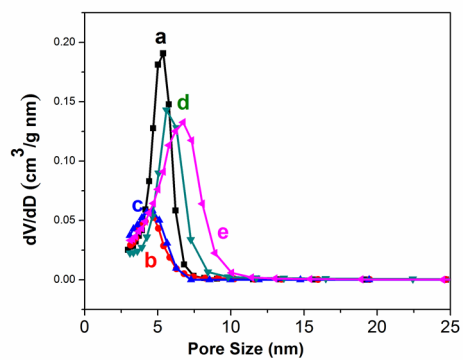
<sup>c</sup>Nitrogen content determined by elemental analysis.

<sup>d</sup>Content of Mo<sub>2</sub>C determined by XPS.

<sup>e</sup>Formaldehyde was used as monomer instead of terephthalaldehyde.

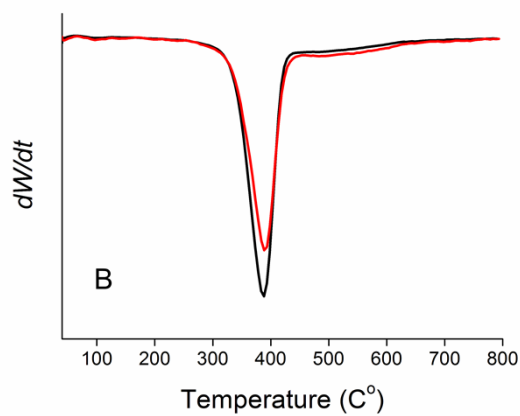
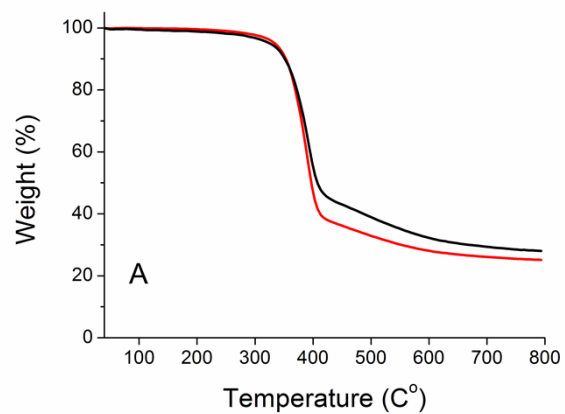


**Figure S1.** SAXS patterns of (A) as-made H-OMC and H-OMC, (b) as-made C-OMC and C-OMC. The patterns were acquired on a Nanostar U small-angle X-ray scattering system using Cu K $\alpha$  radiation.

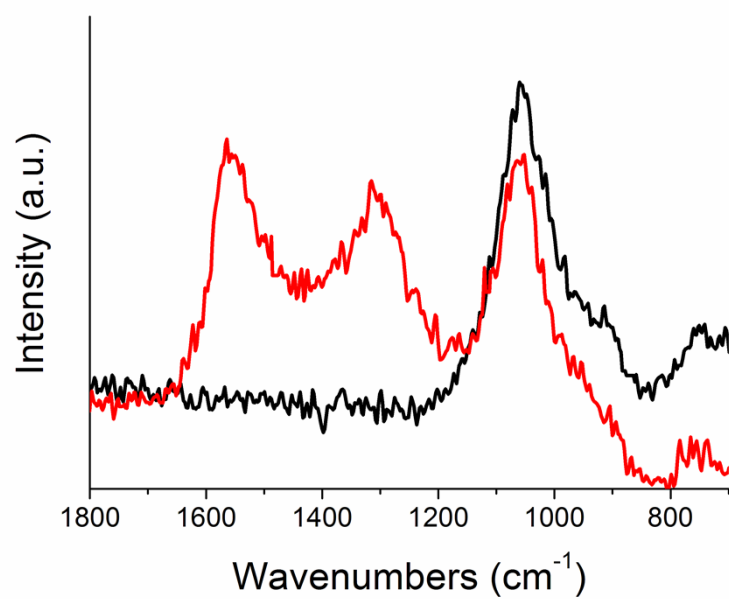


**Figure S2.** BJH pore size distribution of H-OMC (a), C-OMC (b), N-OMC (c), Mo-OMC (d), and F-OMC (e)

prepared via solvent-free synthesis method.



**Figure S3.** TG (A) and DTG (B) curves for H-OMC (black line) and C-OMC (red line). The measurements were carried out on a Mettler Toledo TGA/SDTA851 analyzer from 40°C to 800°C under argon with a rate of 5°C min<sup>-1</sup>.



**Figure S4.** Raman spectra of H-OMC (red line) and background of glass slide (black line). The Raman spectra showed two broad band at 1317 and 1562  $\text{cm}^{-1}$ , assigned to the D band and G band, respectively, suggesting an amorphous carbon framework.

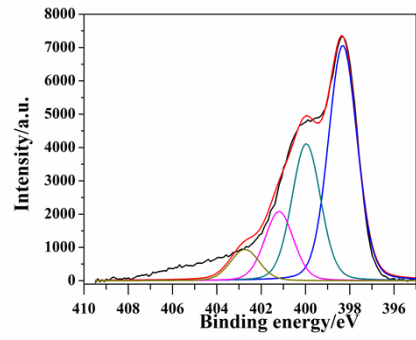
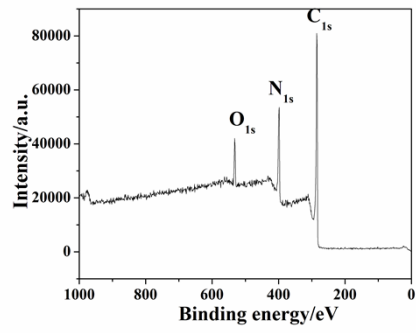
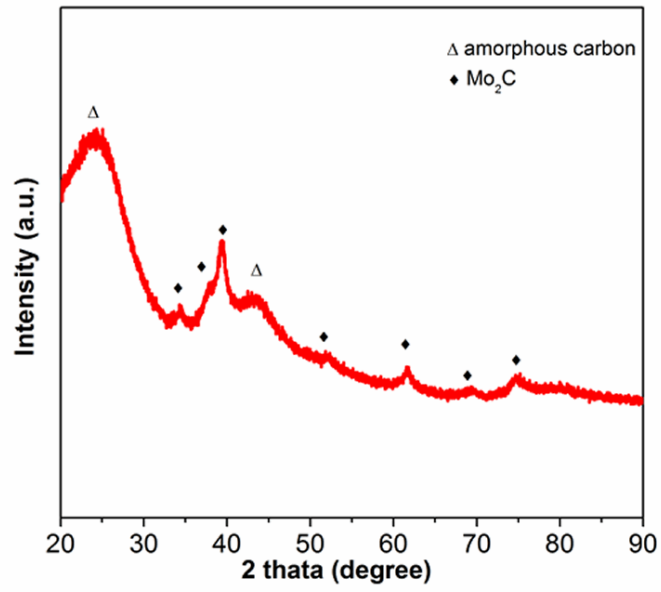


Figure S5. XPS spectra of N-OMC.





**Figure S6.** Wide-angle X-ray diffraction patterns of nanocomposites of Mo-OMC.

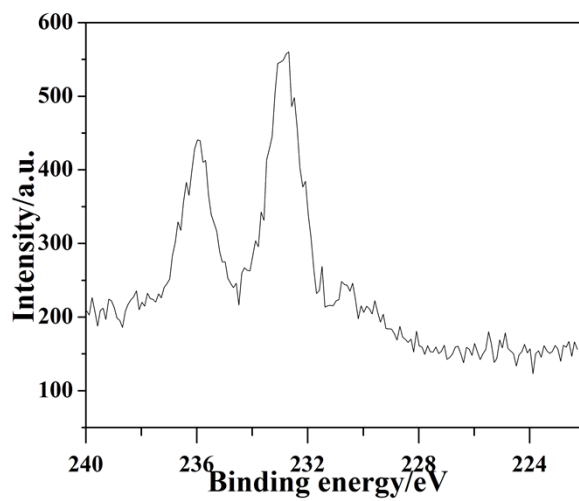
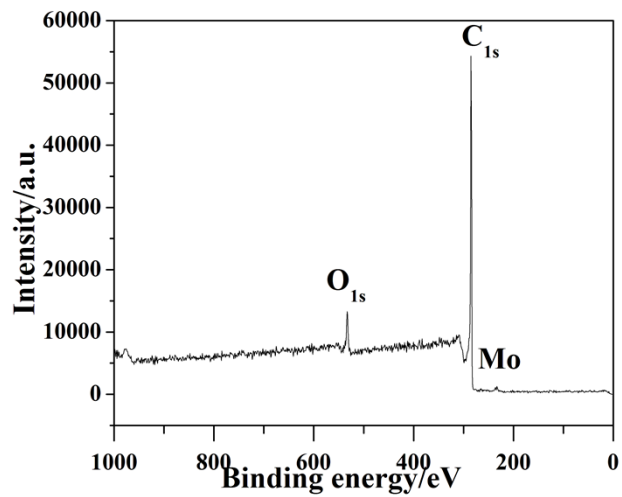
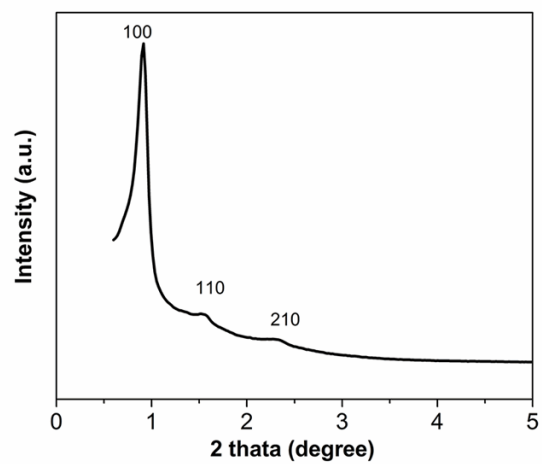


Figure S7. XPS spectra of Mo-OMC.



**Figure S8.** Small angle XRD pattern of H-OMC with a large amount of more than 10 g in one-pot

