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 (Ø 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\textalpha radiation (40 kV, 40 mA). The d-spacing values were calculated using the Braggs diffraction formula of 2d\textalpha sinθ.
\[ n = \lambda, \text{ and the unit parameters } \lambda = 2d_{10}^{1/2} \text{ for OMC and } \lambda = d_{110}^{1/2} \text{ for cubic carbon of C-OMC.} \]

The \( \text{N}_2 \) 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, \( t/\text{Å} = \left[13.99/(\log(P_0/P) + 0.0340)\right]^{1/2} \). 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

<table>
<thead>
<tr>
<th>Sample</th>
<th>F127/g</th>
<th>Resorcinol/g</th>
<th>Terephthalaldehyde/g</th>
<th>Melamine/g</th>
<th>Mo source/g</th>
</tr>
</thead>
<tbody>
<tr>
<td>H-OMC</td>
<td>0.62</td>
<td>0.22</td>
<td>0.40</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>C-OMC</td>
<td>0.41</td>
<td>0.22</td>
<td>0.40</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>N-OMC</td>
<td>1.50</td>
<td>0.44</td>
<td>0.56</td>
<td>2.0</td>
<td>-</td>
</tr>
<tr>
<td>Mo-OMC</td>
<td>0.62</td>
<td>0.22</td>
<td>0.40</td>
<td>-</td>
<td>0.030</td>
</tr>
<tr>
<td>F-OMC</td>
<td>0.62</td>
<td>0.44</td>
<td>0.49</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

*a Melamine served as N source.

*b Phosphomolybdic acid served as Mo source and the nominal content of Mo was ~5 wt%.

*c Nitrogen content determined by elemental analysis.

*d Content of MoC determined by XPS.

*e 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 Kα 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⁻¹.
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 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.