Supporting Information

Multi-template synthesis of hierarchical porous carbon spheres

with potential application in supercapacitors

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Fig. S1 SEM images of pristine CaCO₃ rods.



Fig. S2 Optical micrographs of $CaCO_3$ -rod stabilised w/o emulsions and $CaCO_3$ /silica composite capsules prepared with different APTES contents. a) 0.04 mL APTES, $CaCO_3$ -rod stabilised emulsions (a1), the emulsion aged at 60 °C for 20 h (a2), and $CaCO_3$ /silica composite capsules (a3). b) 0.10 mL APTES. c) 0.20 mL APTES. d) 0.40 mL APTES. e) 0.80 mL APTES. f) 1.20 mL APTES. The inset graph of each shows a single emulsion droplet or capsule.



Fig. S3 Optical micrographs of silica capsules prepared with different APTES contents. a) 0.04 mL APTES, wet and dried silica capsules. b) 0.10 mL APTES. c) 0.20 mL APTES. d) 0.40 mL APTES. e) 0.80 mL APTES. f) 1.20 mL APTES. The inset graph of each shows a single silica capsule.



Fig. S4 Small angle X-ray diffraction (XRD) patterns of silica capsules prepared with different APTES contents. a) 0.04 mL APTES. b) 0.10 mL APTES. c) 0.20 mL APTES. d) 0.40 mL APTES. e) 0.80 mL APTES. f) 1.20 mL APTES.



Fig. S5 Influence of no addition of polyvinyl pyrrolidone (PVP): Deionized water was used to replace 2.5 wt% PVP aqueous solution as the inner phase of CaCO₃-rod stabilised w/o emulsions. Optical micrographs of CaCO₃-rod stabilised w/o emulsions and CaCO₃/silica composite capsules prepared with different APTES contents. a) 0.04 mL APTES, CaCO₃-rod stabilised emulsions (a1), the emulsion aged at 60 °C for 20 h (a2), and CaCO₃/silica composite capsules (a3). b) 0.10 mL APTES. c) 0.20 mL APTES. d) 0.40 mL APTES. e) 0.80 mL APTES. f) 1.20 mL APTES. The inset graph of each shows a single emulsion droplet or capsule.



Fig. S6 Influence of no addition of polyvinyl pyrrolidone (PVP): Deionized water was used to replace 2.5 wt% PVP aqueous solution as the inner phase of CaCO₃-rod stabilised w/o emulsions. Optical micrographs of silica capsules prepared with different APTES contents. a) 0.04 mL APTES, wet and dried silica capsules. b) 0.10 mL APTES. c) 0.20 mL APTES. d) 0.40 mL APTES. e) 0.80 mL APTES. f) 1.20 mL APTES. The inset graph of each shows a single silica capsule.



Fig. S7 Influence of no addition of polyvinyl pyrrolidone (PVP): Deionized water was used to replace 2.5 wt% PVP aqueous solution as the inner phase of $CaCO_3$ -rod stabilised w/o emulsions. Microstructure evolution of hierarchical silica capsules with the increase of APTES content in the oil phase, observed by Cryo-SEM. a) 0.04 mL APTES. b) 0.10 mL APTES. c) 0.20 mL APTES. d) 0.40 mL APTES. e) 0.80 mL APTES. f) 1.20 mL APTES. Scale bars, 50 µm for a1, b1, c1, d1, e1 and f1; 5 µm for a2, b2, c2, d2, e2 and f2; 0.5 µm for a3, b3, c3, d3, e3 and f3.



Fig. S8 SEM images of hierarchical porous carbon (HPC) spheres. a) HPC1 prepared from the hierarchical silica based on the system with 0.40 mL APTES. b) HPC2 prepared from the hierarchical silica based on the system with 0.80 mL APTES. c) HPC3 prepared from the hierarchical silica based on the system with 1.20 mL APTES. Scale bars, 100 μ m for a1, b1 and c1; 20 μ m for a2, b2 and c2.



Fig. S9 Small angle XRD patterns of hierarchical porous carbon (HPC) spheres. a) HPC1 prepared from the hierarchical silica based on the system with 0.40 mL APTES. b) HPC2 prepared from the hierarchical silica based on the system with 0.80 mL APTES. c) HPC3 prepared from the hierarchical silica based on the system with 1.20 mL APTES.



Fig. S10 Digital photos of ordered mesoporous polymer composites (OMPC). a) OMPC1 prepared from the original system with 0.40 mL APTES. b) OMPC2 prepared from the original system with 0.80 mL APTES. c) OMPC3 prepared from the original system with 1.20 mL APTES. Scale bars, 1.0 cm for a, b and c.



Fig. S11 A typical Raman spectrum of HPCs (using that of HPC2 as an example).



Fig. S12 CV curves of HPC2 at different scan rates.

Tables

Table	e S1 BET surface	area (S _{bet}), I	Micropore are	ea (S_m) and	d Eterna	al surface	area ((S_e) of S_e	Silica
capsules prepared from the systems with different APTES contents									
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	S_{bet} $[m^2g^{-1}]$	S_m [m ² g ⁻¹]	<i>S_e</i> [m ² g ⁻¹]
Silica capsule 1, 0.04 mL APTES	287	67	220
Silica capsule 2, 0.10 mL APTES	250	55	195
Silica capsule 3,0.20 mL APTES	129	61	68
Silica capsule 4, 0.40 mL APTES	162	42	120
Silica capsule 5, 0.80 mL APTES	206	0	206
Silica capsule 6, 1.20 mL APTES	422	0	422

Table S2 BET surface area (S_{bel}) and Total pore volume (V_l) of hierarchical porous carbons (HPCs) prepared from hierarchical silica based on the systems with different APTES contents

	S _{bet}	V_t
	[m ² g ⁻¹] ^[a]	[m ³ g ⁻¹] ^[b]
HPC1, 0.40 mL APTES	408	0.28
HPC2, 0.80 mL APTES	628	0.46
HPC3, 1.20 mL APTES	759	0.56