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Supporting Information

Interfacial Charge Shielding Directs Synthesis of Dendritic Mesoporous Silica Nanospheres by a Dual-Templating Approach

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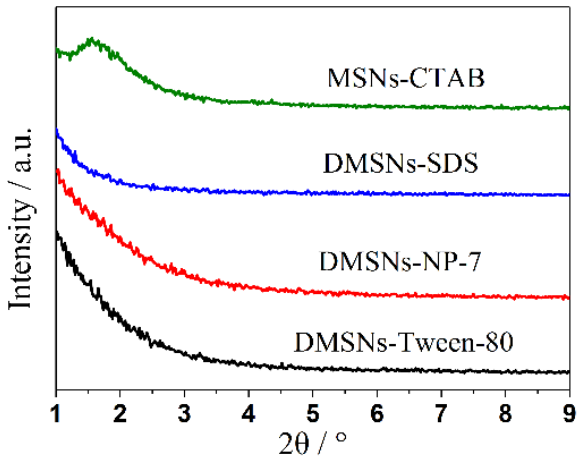

Figure S1 XRD patterns of MSNs synthesized by using different templates.

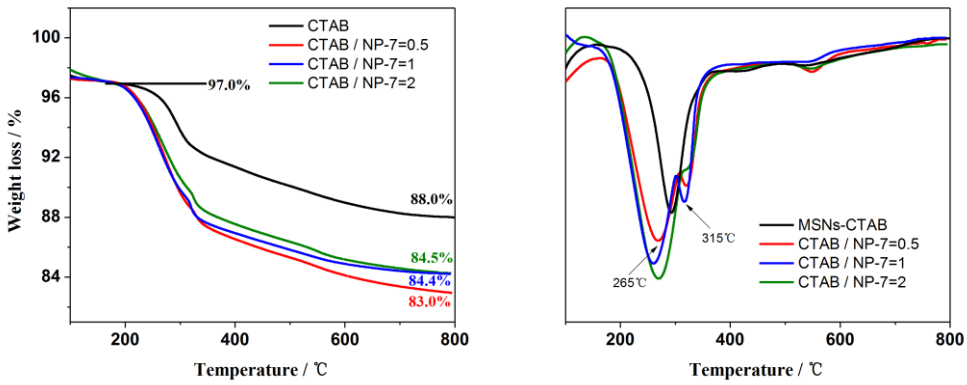

Figure S2. Thermogravimetric analysis (TG) of DMSNs synthesized by using dual templates

CTAB and NP-7 with different molar ratios of CTAB / NP-7 (0.5, 1 and 2) and the TG of MSNs synthesized by using a solo cationic surfactant CTAB (MSNs-CTAB) was demonstrated for comparison. The weight loss centered at 265 °C was attributed to the electrostatically interacting CTA^+ from CTAB, the peak minima centered at 315 °C was due to the incorporation of nonionic surfactant NP-7.

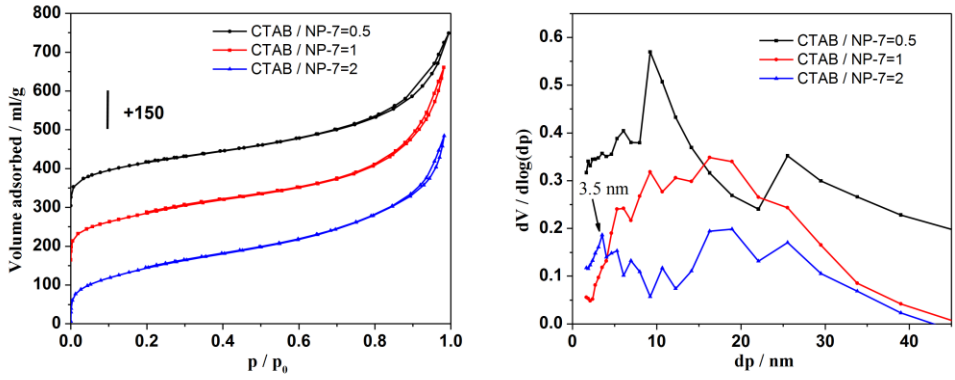

Figure S3. N_2 adsorption-desorption isotherms (left) and pore size distribution (PSD) plots (right) of the DMSNs synthesized by using dual templates CTAB and NP-7 with different molar ratios of CTAB / NP-7: (a) 0.5; (b) 1 and (c) 2, respectively. PSD calculated by the BJH method from desorption branches.

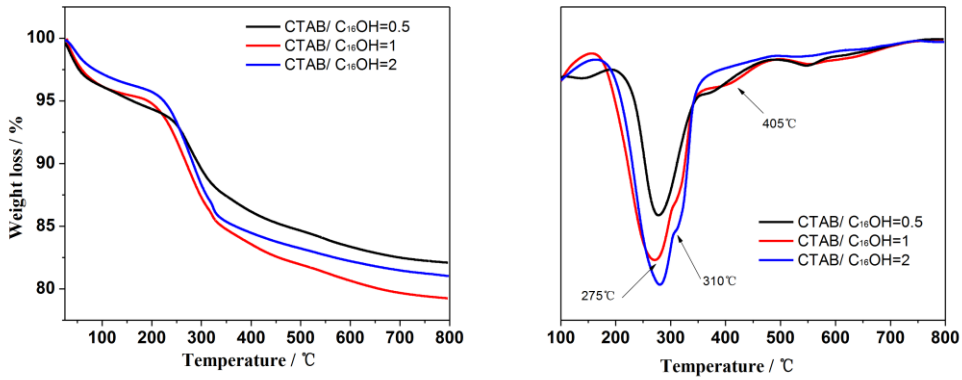

Figure S4. Thermogravimetric analysis (TG) of MSNs synthesized by using dual templates CTAB and $C_{16}OH$ with different molar ratios of CTAB /$C_{16}OH$ (0.5, 1 and 2). The weight loss centered at 275 °C was attributed to the electrostatically interacting CTA^+, the weight loss centered at 310 °C was attributed to the non-interacting CTAB, and the peak minima centered at 405 °C was due to the incorporation of fatty alcohol 1-hexadecanol ($C_{16}OH$).

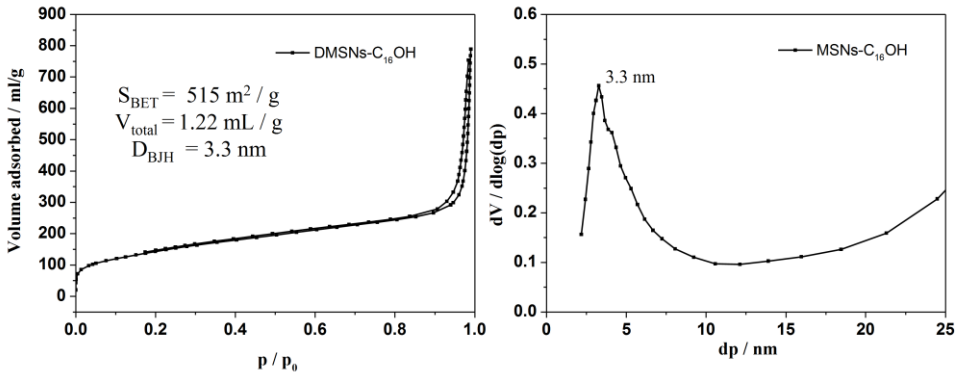

Figure S5. N_2 adsorption−desorption isotherms (left) and pore size distribution (PSD) plots (right) of MSNs synthesized by using templates CTAB and pore swelling agent $C_{16}OH$. PSD calculated by the BJH method from desorption branches.

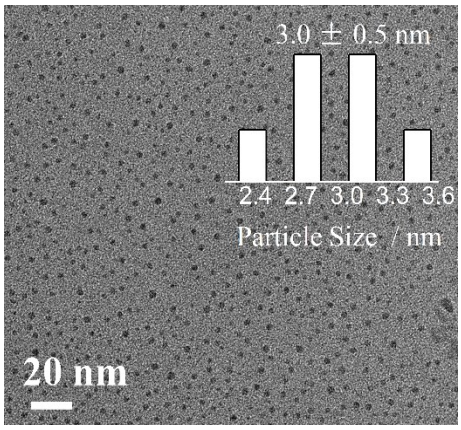

Figure S6 TEM images of MSNs-CTAB samples collected at the reaction time of 30 s.

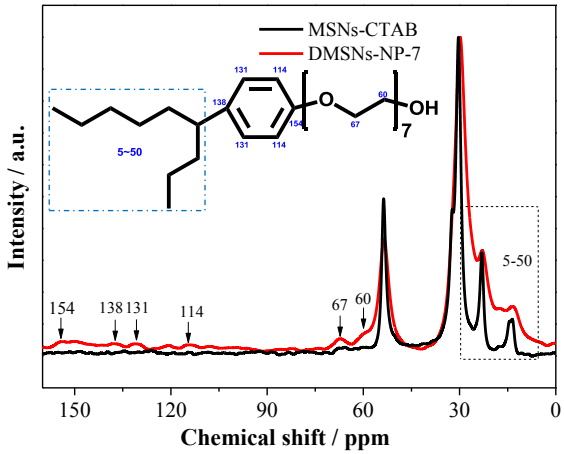

Figure S7. ^{13}C CP MAS NMR spectra of MSNs synthesized at various reaction conditions. Black line represents the MSNs synthesized by using a solo cationic surfactant CTAB, and the red line represents the DMSNs synthesized by using dual templates CTAB and NP-7. Black arrows point out characteristic chemical shifts of nonionic surfactants NP-7.

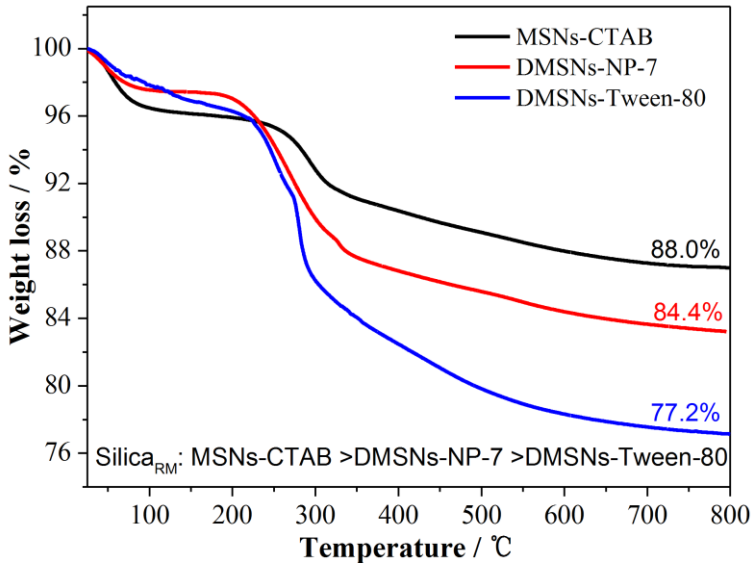

Figure S8. Thermogravimetric analysis (TG) of DMSNs synthesized by using a solo cationic surfactant CTAB and dual templates CTAB with NP-7 or Tween-80, respectively. Silica$_{RM}$ means silica residual mass in unit of percentage obtained from TG analysis at 800 ℃.

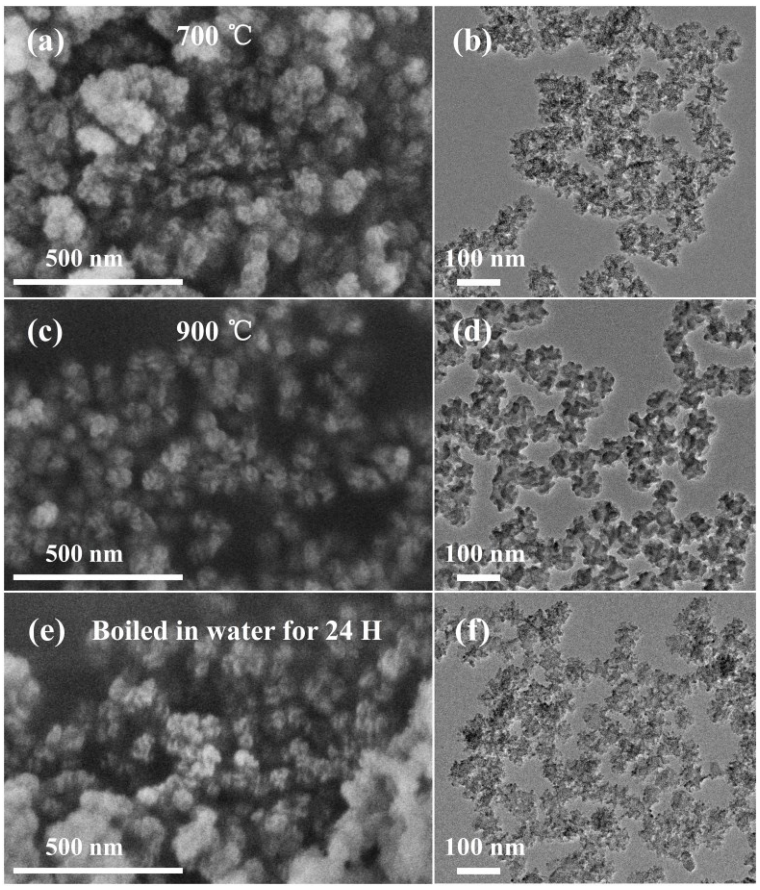

Figure S9. SEM (left) and TEM (right) images of DMSNs-NP-7 calcined at varied temperatures of 700 ℃ (a and b), 900 ℃ (c and d), and boiled in water for 24 hours (e and f).

Table S1. Textural Characteristics of Calcined MSNs Synthesized in Various Reaction Conditions

Sample[a]	S_{BET}[b] (m^2 / g)	V_{total}[c] (ml / g)	V_{Meso}[d] (ml / g)	D_{BJH}[e] (nm)	PSD[f] (nm)
MSNs	675	1.25	0.43	2.4	50 ± 10
DMSNs-Tween-80	371	0.59	0.28	16	74 ± 10
DMSNs-NP-7-0.5[g]	474	0.71	0.38	12	74±10
DMSNs-NP-7-1	502	0.80	0.41	15	74±10
DMSNs-NP-7-2	565	0.77	0.45	3.5/16	67±10
MSNs-C_{16}OH	515	1.22	0.38	3.3	74±10
DMSNs-CTATos[h]	552	1.45	0.41	3.2/16	120 ± 10
DMSNs-SDS[i]	450	1.59	0.83	3.2/10.6	120 ± 10
MCM-41[j]	853	0.79	0.70	2.9	-

[a]MSNs were synthesized by a single surfactant CTAB or CTAB with C_{16}OH, denoted as MSNs-CTAB and MSNs-C_{16}OH. [a]DMSNs were synthesized by CTAB with NP-7, Tween-80, and SDS, or a single surfactant CTATos, denoted as DMSNs-NP-7 , DMSNs-Tween-80, DMSNs-SDS and DMSNs-CTATos, respectively. [b]Specific surface area measured from N_2 physisorption. [c]Total pore volume measured at P/P_0= 0.99. [d]Meso pore volume measured at P/P_0= 0.80. [e]Pore diameter calculated from the BJH theoretical model. [f]Particle size distribution was determined by measuring the diameters of at least 100 particles under TEM. [g]DMSNs-NP-7-X, X represented the CTAB/NP-7 molar ratio. [h]From ref 1. [i]From ref 2. [j]From ref 3.

Table S2. Formation of cyclohexane oxide (CHO) from cyclohexene using different catalysts[a]

Catalyst	Conv/%	CHO sel/%	CHO yield/%
Ti-MCM-41-Cal[b]	25.7	92.2	23.7
Ti-MCM-41-Sil	22.8	97.0	22.1
Ti-DMSNs-Cal	14.8	95.5	14.1
Ti-DMSNs-Sil	24.5	96.0	23.5

[a]Reaction conditions: Catalyst, 50 mg; acetonitrile, 10 ml; cyclohexene, 10 mmol; TBHP (5.5 M in decane), 10 mmol; temp., 333 K; time, 2 h. [b]Ti-MCM-41-X, X represented the remove methods of templates: calcination (Cal) or trimethylsilylation (Sil). It is same to Ti-DMSNs.

References

1. K. Zhang, L. L. Xu, J. G. Jiang, N. Calin, K. F. Lam, S. J. Zhang, H. H. Wu, G. D. Wu, B. Albela, L. Bonneviot and P. Wu, *Journal of the American Chemical Society*, 2013, **135**, 2427-2430.
2. P. C. Liu, Y. J. Yu, B. Peng, S. Y. Ma, T. Y. Ning, B. Q. Shan, T. Q. Yang, Q. S. Xue, K. Zhang and P. Wu, *Green Chemistry*, 2017, **19**, 5575-5581.
3. K. Zhang, H.-L. Chen, B. Albela, J.-G. Jiang, Y.-M. Wang, M.-Y. He and L. Bonneviot, *European Journal of Inorganic Chemistry*, 2011, **2011**, 59-67.