Supplementary Information

Temperature-regulated construction of hierarchical titanosilicate zeolites

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Supplementary Figures and Tables

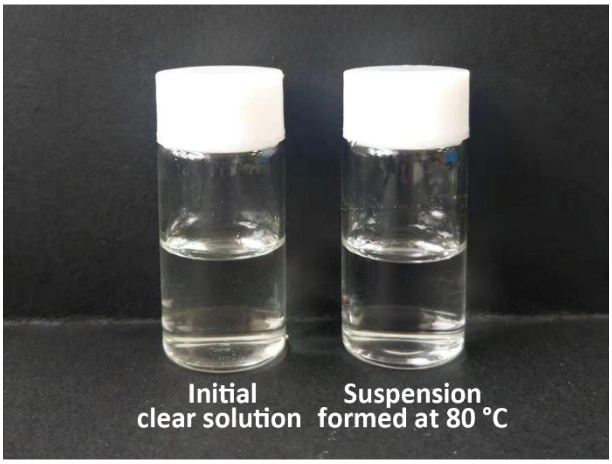

Figure S1. Photographs of the initial clear solution and the suspension formed at 80 °C for 1 day.

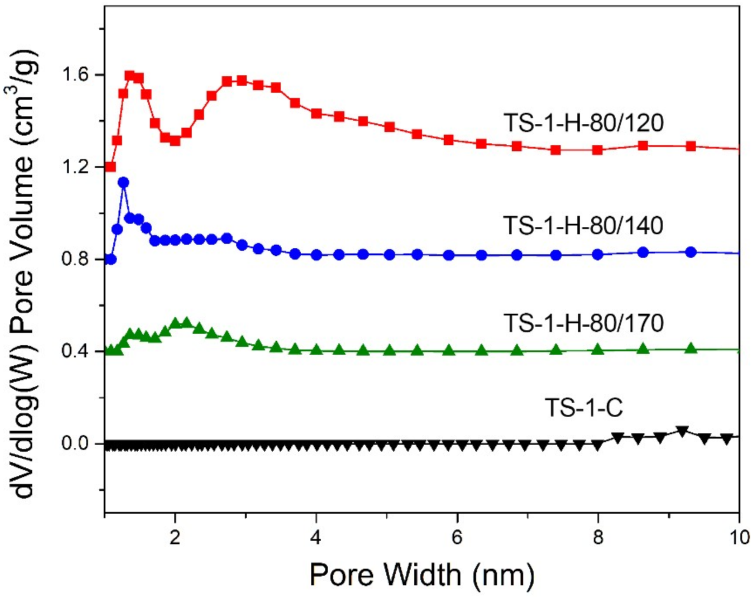

Figure S2. Pore size distributions of TS-1-H-80/120, TS-1-H-80/140, TS-1-H-80/170, and TS-1-C samples.

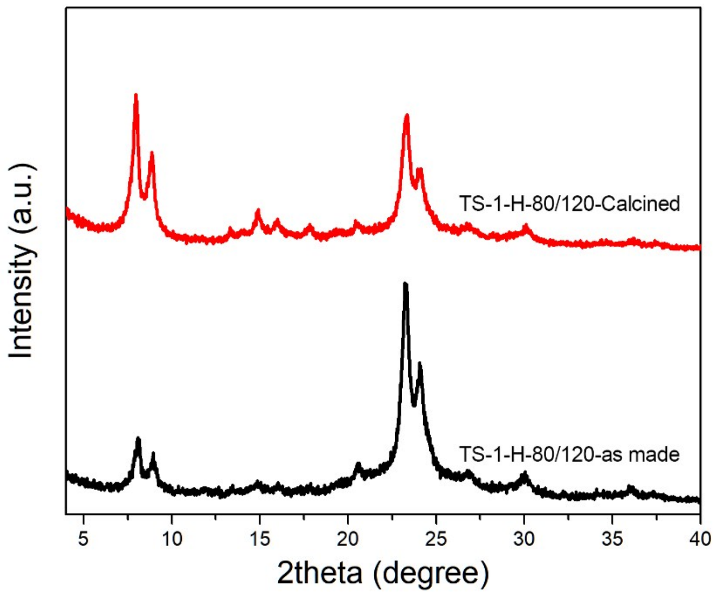

Figure S3. XRD patterns of as-made TS-1-H-80/120 sample and the corresponding calcined sample.

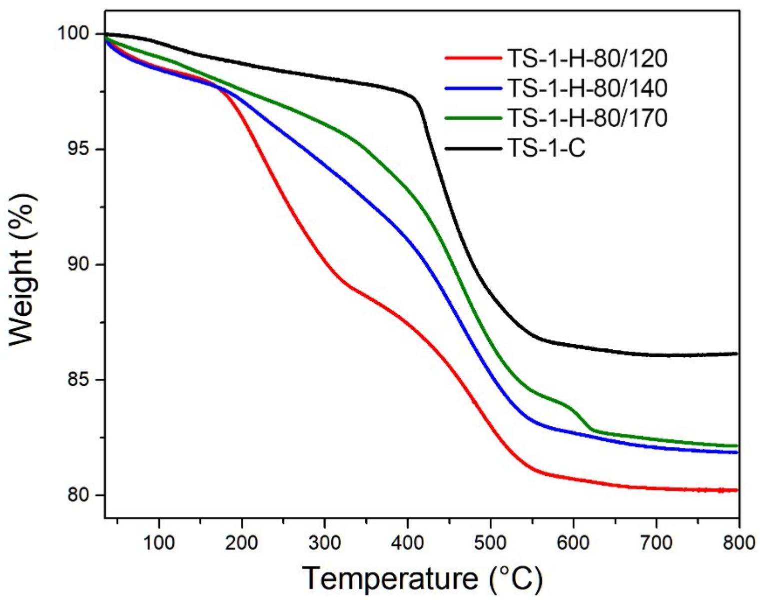

Figure S4. Thermogravimetric (TG) curves of the as-synthesized TS-1-H-80/120, TS-1-H-80/140, TS-1-H-80/170, and TS-1-C samples.

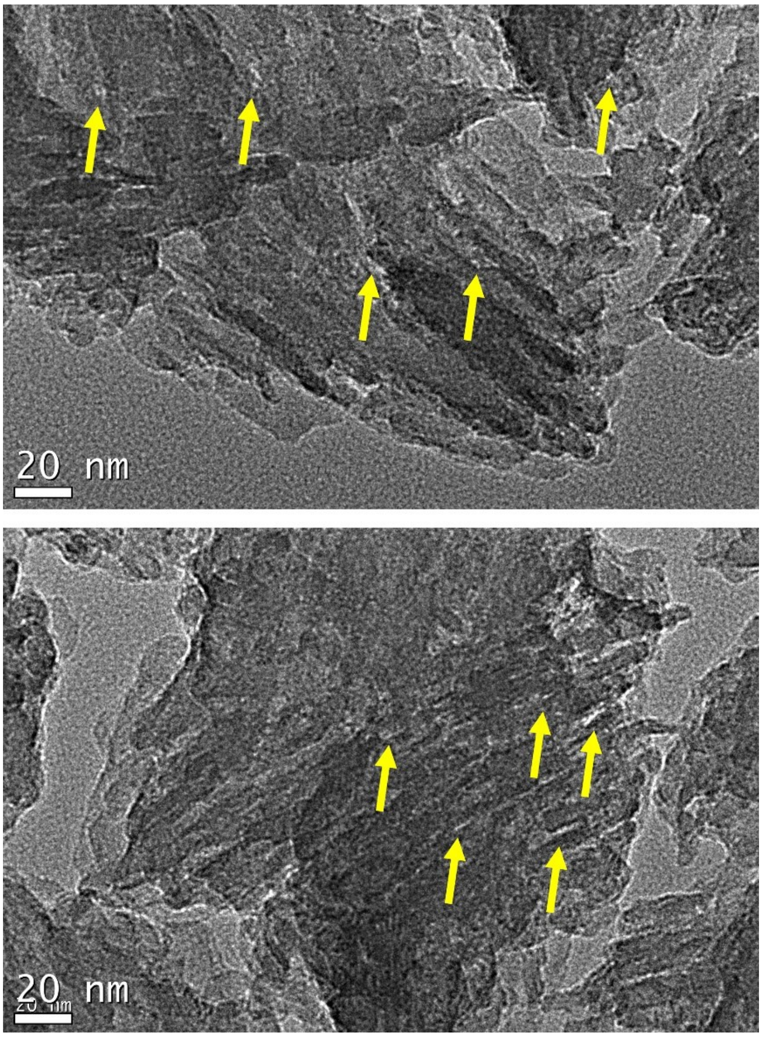

Figure S5. Higher-resolution transmission electron microscopy (HRTEM) images of the TS-1-H-80/120 sample.

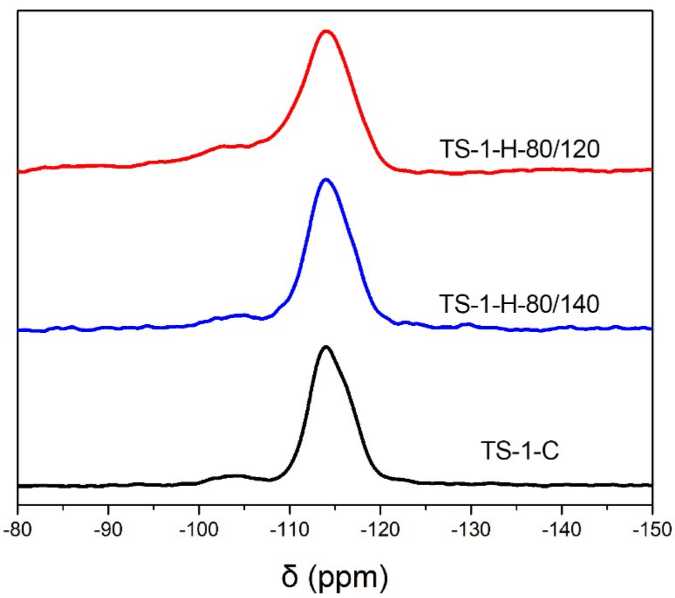

Figure S6. ^{29}Si MAS NMR spectra of TS-1-H-80/120, TS-1-H-80/140, and TS-1-C samples.

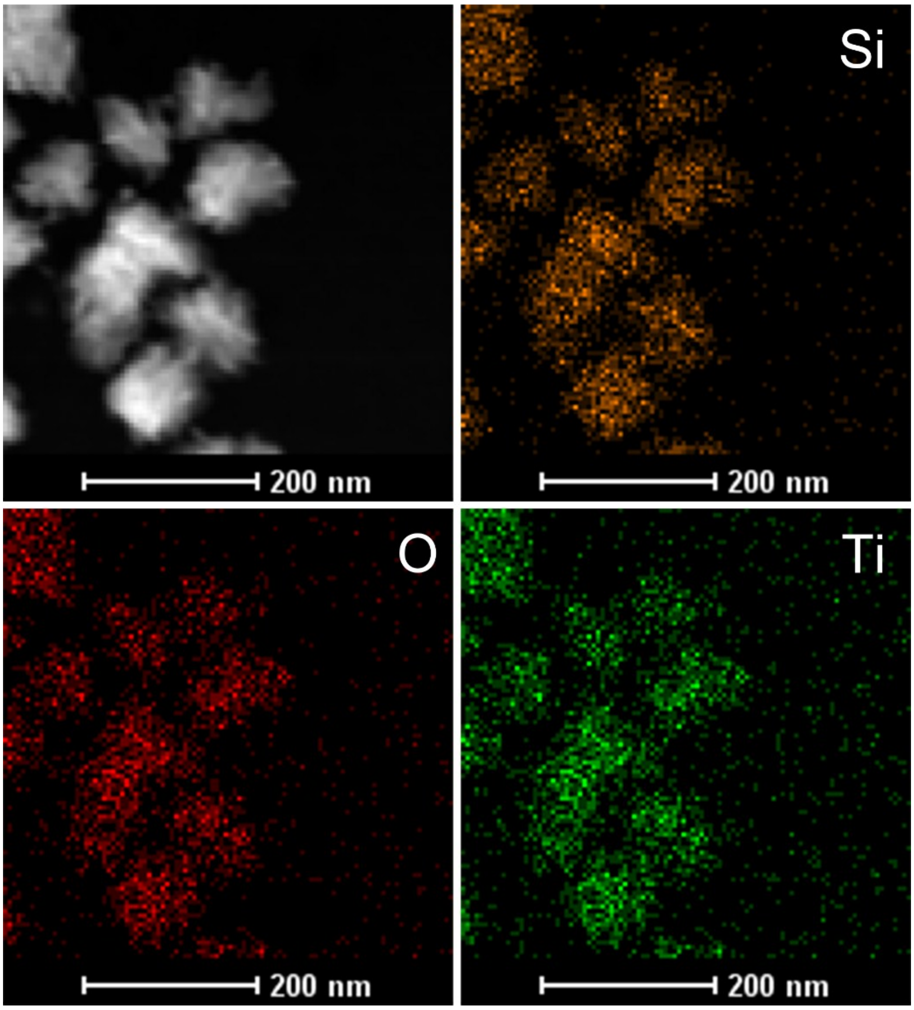

Figure S7. High-angle annular dark field scanning transmission electron microscopy (HAADF STEM) image and elemental mappings for Si, O, and Ti elements of TS-1-H-80/120 sample.

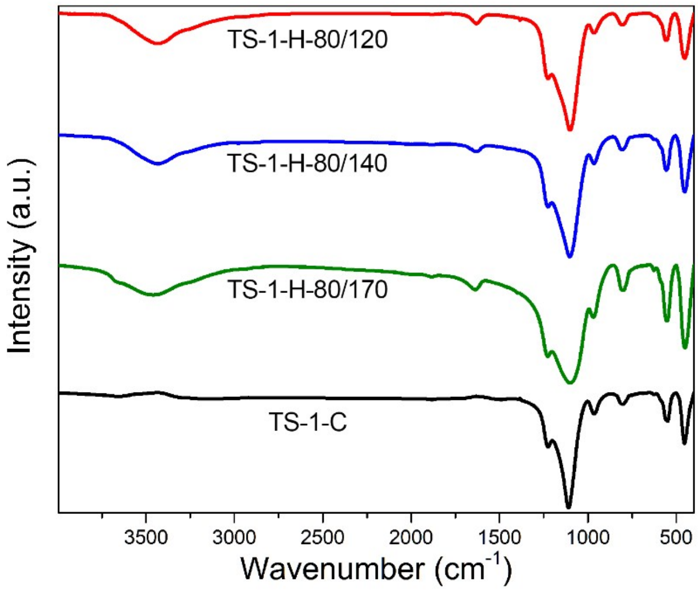

Figure S8. FT-IR spectra of TS-1-H-80/120, TS-1-H-80/140, TS-1-H-80/170, and TS-1-C samples.

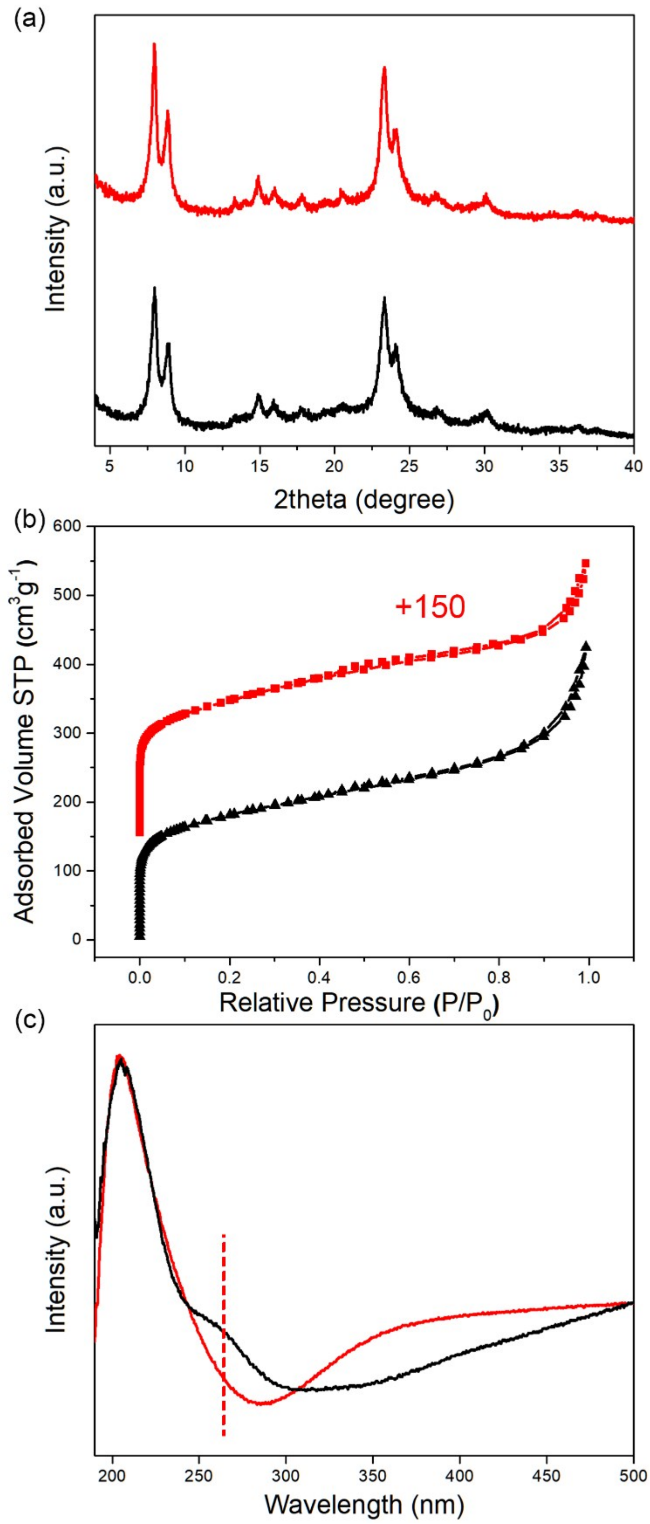

Figure S9. XRD pattern (a), N_2 adsorption and desorption isotherm (b), and UV-vis spectrum (c) of the fresh TS-1-H-80/120 sample (red line) and the sample after ten cycles in the oxidation of DBT (black line).

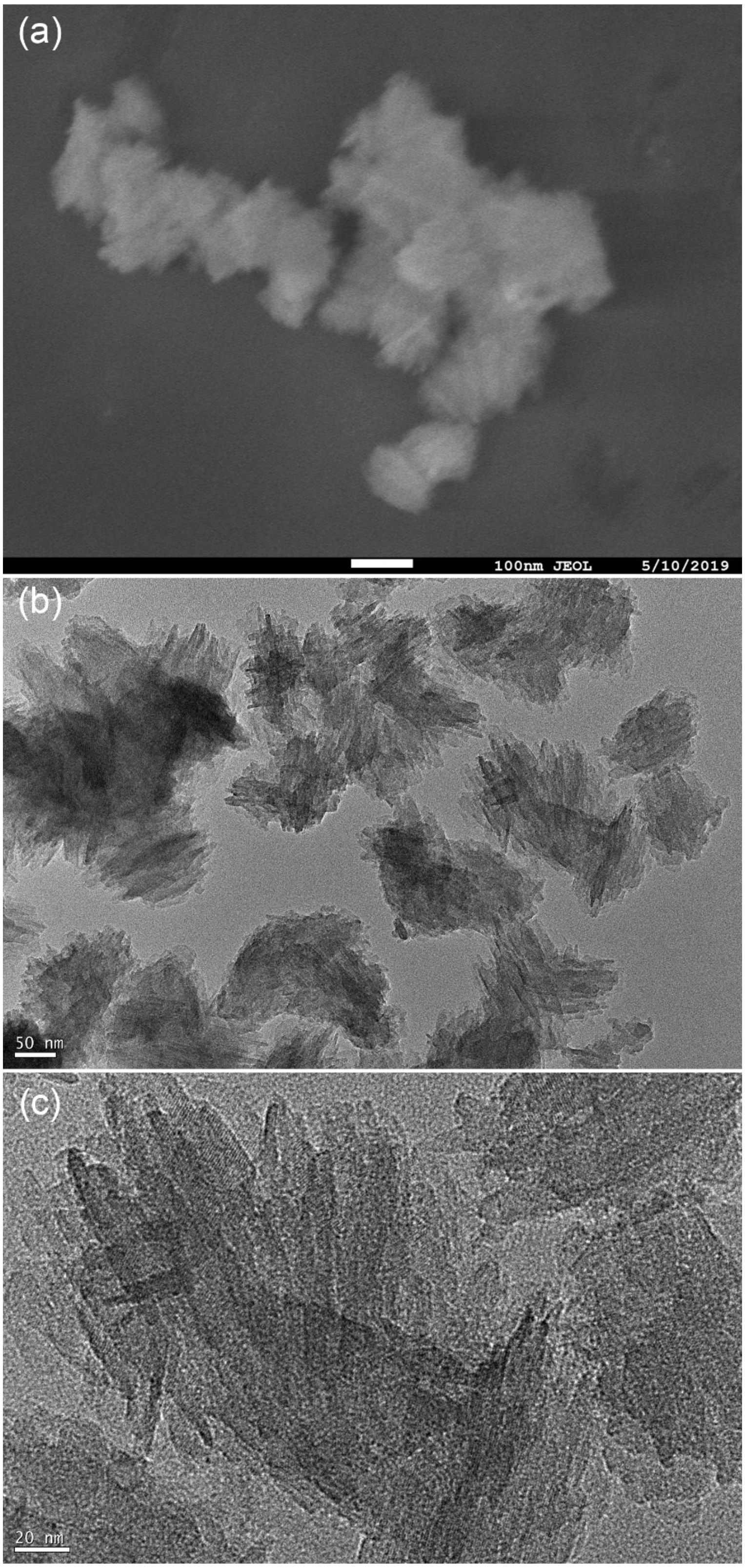

Figure S10. SEM (a) and TEM (b, c) images of the TS-1-H-80/120 sample after ten cycles in the oxidation of DBT.

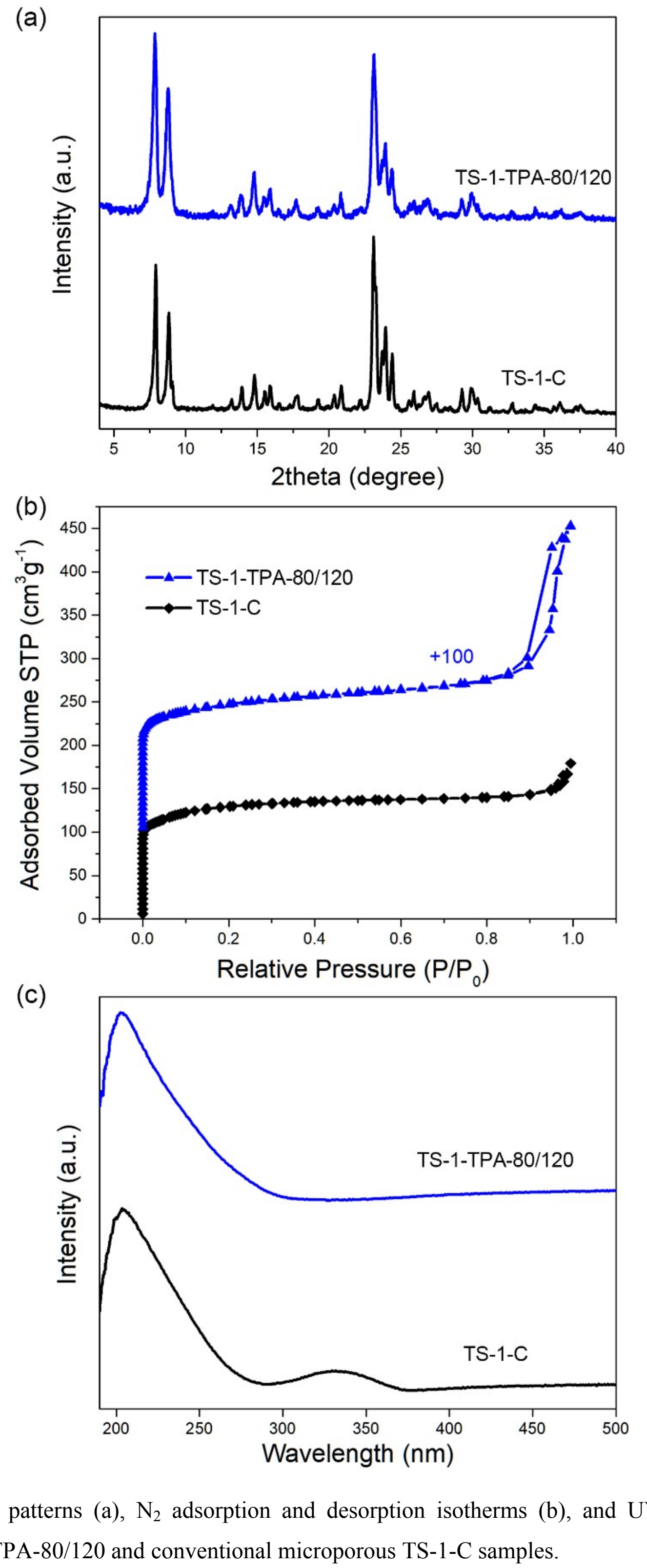

Figure S11. XRD patterns (a), N_2 adsorption and desorption isotherms (b), and UV-Vis spectra (c) of the hierarchical TS-1-TPA-80/120 and conventional microporous TS-1-C samples.

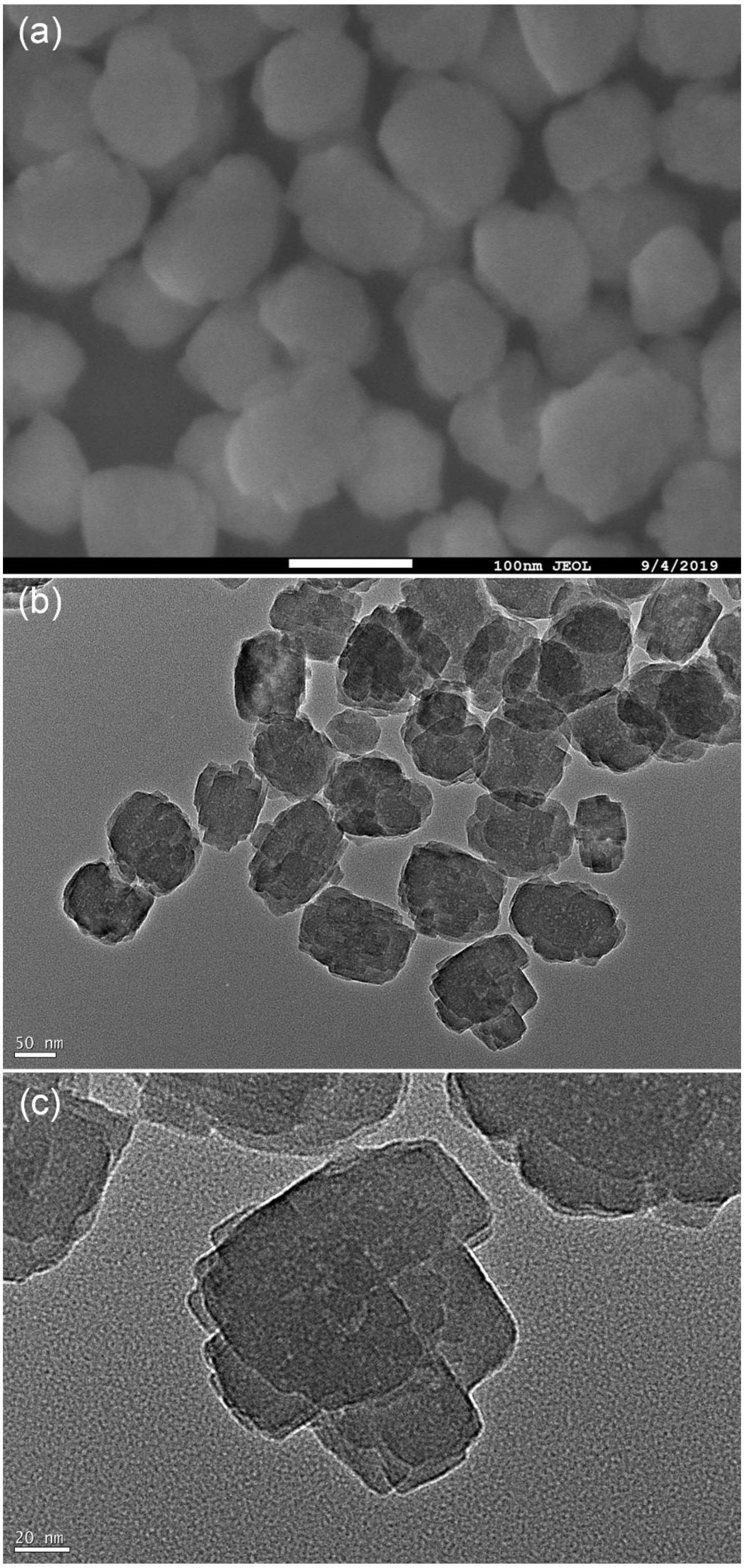

Figure S12. SEM (a) and TEM (b, c) images of hierarchical TS-1-TPA-80/120 sample.

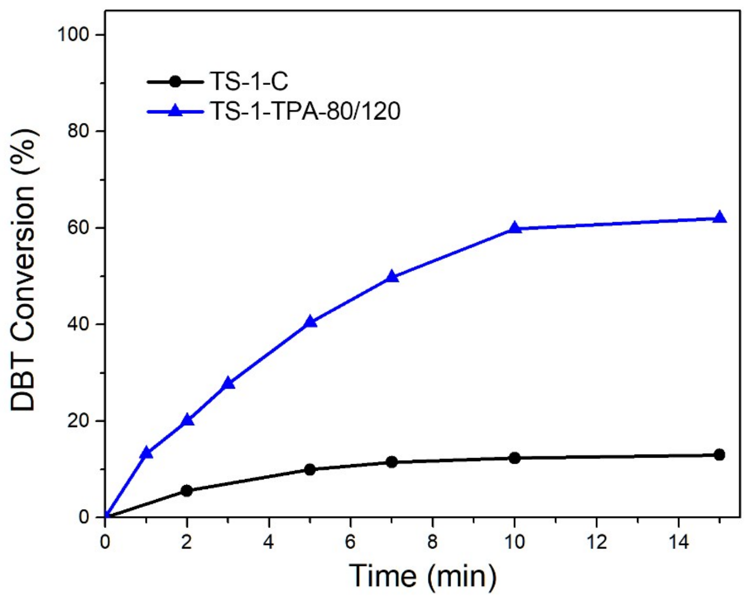

Figure S13. Catalytic oxidation of DBT with TBHP over microporous TS-1-C and hierarchical TS-1-TPA-80/120 samples. Reaction conditions: 10mL 500ppm model fuels, 60 °C, 30mg catalysts, n(DBT)/n(TBHP) was 0.5.

Table S1. Framework compositions and textural properties of the as-prepared zeolite samples.

Samples	Si/Ti[a]	S_{BET}[b] (m^2/g)	S_{micro}[c] (m^2/g)	S_{ext}[c] (m^2/g)	V_{micro}[c] (cm^3/g)	V_{total}[d] (cm^3/g)	V_{meso}[e] (cm^3/g)
TS-1-H-80/120	106	716	268	448	0.11	0.61	0.50
TS-1-H-80/140	103	597	346	251	0.14	0.44	0.30
TS-1-H-80/170	62	500	353	147	0.14	0.27	0.13
TS-1-C	60	485	310	175	0.12	0.28	0.16
TS-1-TPA-80/120	105	557	368	189	0.15	0.55	0.40
TS-1-H-80/120[f]	191	641	262	379	0.11	0.65	0.54
TS-1-H-120[g]	136	692	280	412	0.12	0.45	0.33

[a] Measured by Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES). [b] Specific surface area calculated from the nitrogen adsorption isotherm using the BET method. [c] S_{micro} (micropore area), S_{ext} (external surface area), and V_{micro} (micropore volume) calculated using the t-plot method. [d] Total pore volume at $P/P_0 = 0.99$. [e] $V_{meso} = V_{total} - V_{micro}$. [f] The sample was used after 10 catalytic reaction-regeneration cycle times. [g] TS-1-H-120 was synthesized with the same molar composition of the TS-1-H-80/120 sample by crystallization at 120 °C for 2 days.

Table S2. Compositional analyses of the as-synthesized samples.

	C (wt.%)	H (wt.%)	N (wt.%)	C/N
TS-1-H-80/120	14.0 [a]	3.1	0.82	19.9 (Calc. 16 [b])
TS-1-H-80/140	12.3	2.6	0.79	18.2 (Calc. 16)
TS-1-H-80/170	11.6	2.4	0.73	18.5 (Calc. 16)
TS-1-C	9.6	2.1	0.82	13.6 (Calc. 12)

[a] Elemental analysis was based on a C, H, N, S elemental analyses. [b] The ideal C/N atomic ratio of the OSDA molecules calculated in their cationic form.

Table S3. Thermogravimetric (TG) analyses of the as-synthesized samples.

		H_2O weight loss (%)	Template weight loss (%)	Template weight loss (%)	Total (wt. %)
TS-1-H-80/120	Temperature (ºC)	35-180	180-320	320-800	19.77
	weight loss (%)	2.62	8.04	9.11	
TS-1-H-80/140	Temperature (ºC)	35-180	180-400	400-800	18.14
	weight loss (%)	2.46	6.48	9.20	
TS-1-H-80/170	Temperature (ºC)	35-180	180-400	400-800	17.86
	weight loss (%)	2.13	4.64	11.09	
TS-1-C	Temperature (ºC)	35-180	180-400	400-800	13.86
	weight loss (%)	1.16	1.49	11.21	