Supporting Information

Sheet-like Silicalite-1 Single Crystals with Embedded Macropores Displaying Superior Catalytic Performance

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Table S1 Precursors' compositions and crystallization conditions

Samples	Precursors' compositions (mol ratio)			Crystallization temperature (K)
	$TPAOH/SiO_2$	H_2O/SiO_2	$urea/SiO_2$	
HMS-S1	0.17	0.9	1.0	423
HMS-S2	0.17	1.8	1.0	423
HMS-Silicalite-1.0	0.17	2.3	1.0	423
HMS-S3	0.17	2.7	1.0	423
HMS-S4	0.17	1.5	0.8	423
HMS-Silicalite-0.8	0.17	1.8	0.8	423
HMS-S5	0.17	2.1	0.8	423
HMS-S6	0.17	0.9	0.5	423
HMS-Silicalite-0.5	0.17	1.5	0.5	423
HMS-S7	0.17	1.8	0.5	423

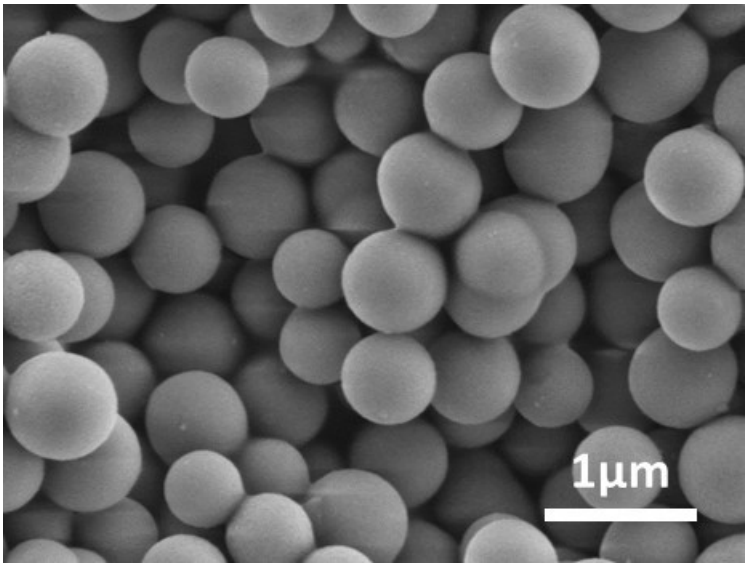

Fig. S1 SEM image of mesoporous silica spheres (MSS)

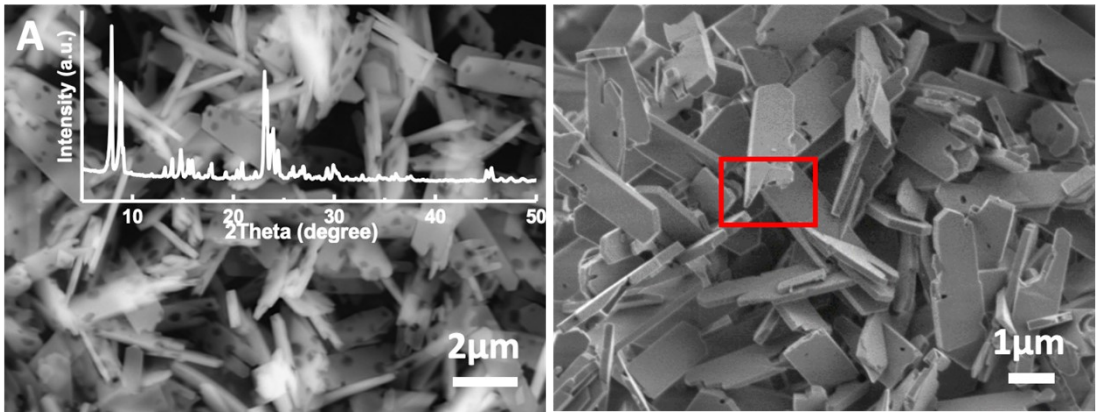

Fig. S2 XRD pattern and SEM image recorded with high (A) and low (B) accelerating voltage of HMS-Silicalite-1-1.0.

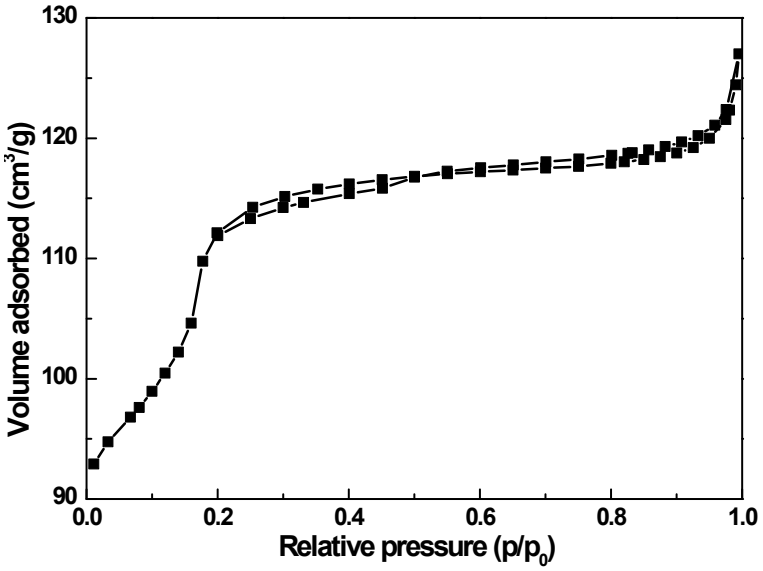

Fig. S3 N_2 adsorption-desorption isotherms of HMS-Silicalite-1-1.0

Table S2 Textural properties of samples HMS-Silicalite-1

Samples	Surface area (m^2/g)			Pore volume (cm^3/g)	
	S_{BET}	S_{mic}	S_{ext}	V_{mic}	V_{total}
HMS-Silicalite-1-1.0	389	283	106	0.115	0.188
HMS-Silicalite-1-0.8	396	294	102	0.116	0.186
HMS-Silicalite-1-0.5	399	293	106	0.122	0.194

[a] S_{BET} (total surface area) calculated by applying BET equation. [b] S_{mic} (micropore area), V_{mico} (micropore volume) and S_{ext} (external surface area) measured by the *t*-plot method. [c] V_{total} (total volume) estimated at relative pressure of 0.99.

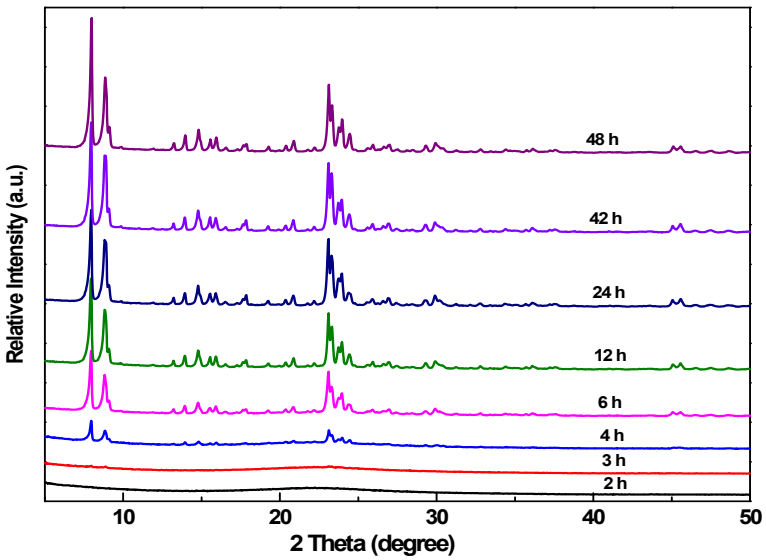

Fig. S4 XRD patterns of the products synthesized from MSS by urea-assisted dry-gel conversion method for different crystallization time, urea/SiO_2 = 1.0

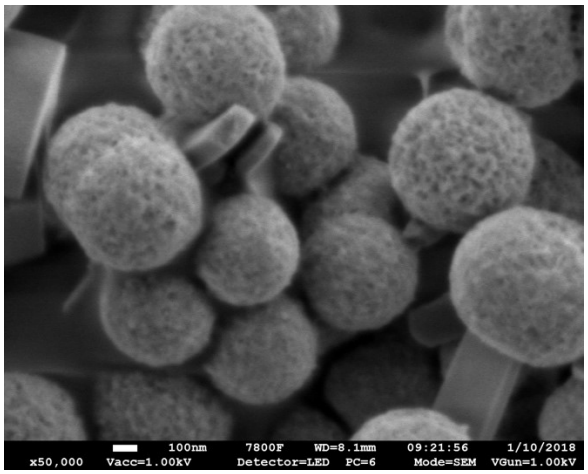

Fig. S5 High-resolution SEM image of the samples synthesized from MSS by urea-assisted dry-gel conversion method for 4 h, image was recorded at voltage of 1 kV

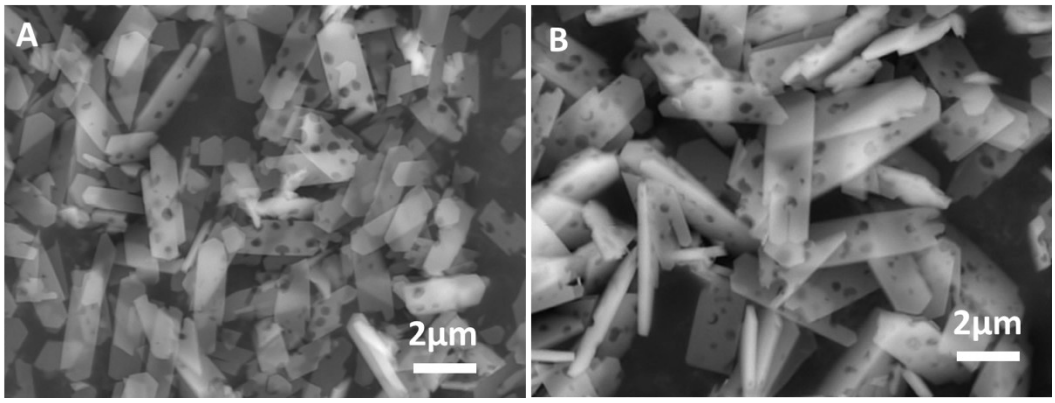

Fig. S6 SEM images of the samples synthesized from MSS by urea-assisted dry-gel conversion method for (A) 12 h and (B) 42 h, images were recorded at voltage of 15 kV

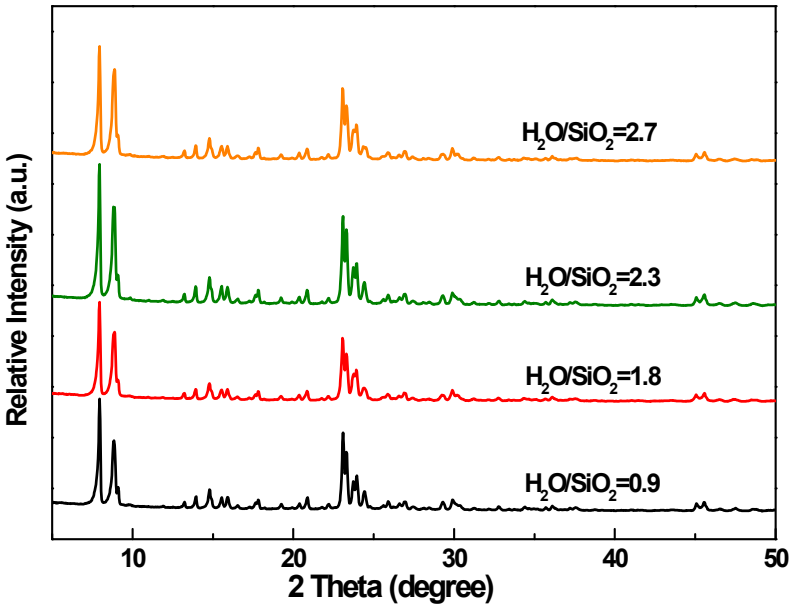

Fig. S7 XRD patterns of samples synthesized with different H_2O/SiO_2 ratios at urea/SiO_2 = 1.0

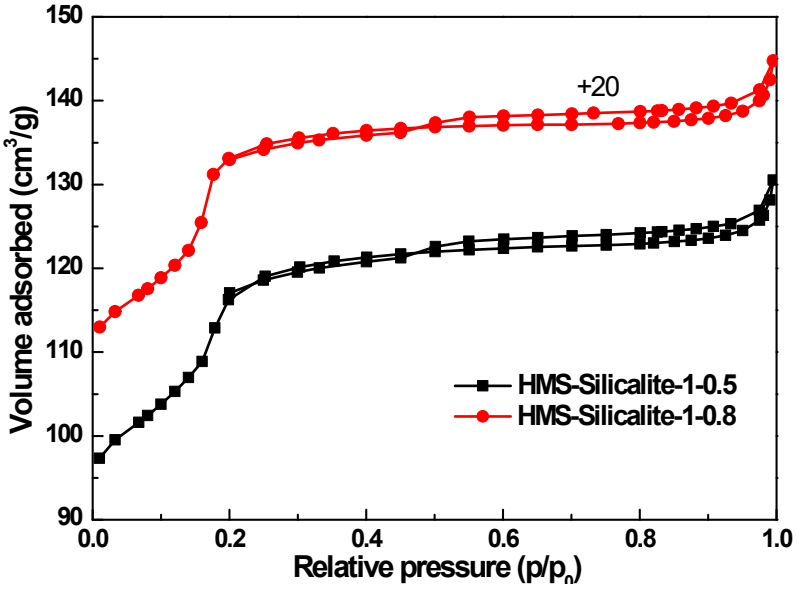

Fig. S8 N_2 adsorption-desorption isotherms of samples HMS-Silicalite-1-0.5 and HMS-Silicalite-1-0.8

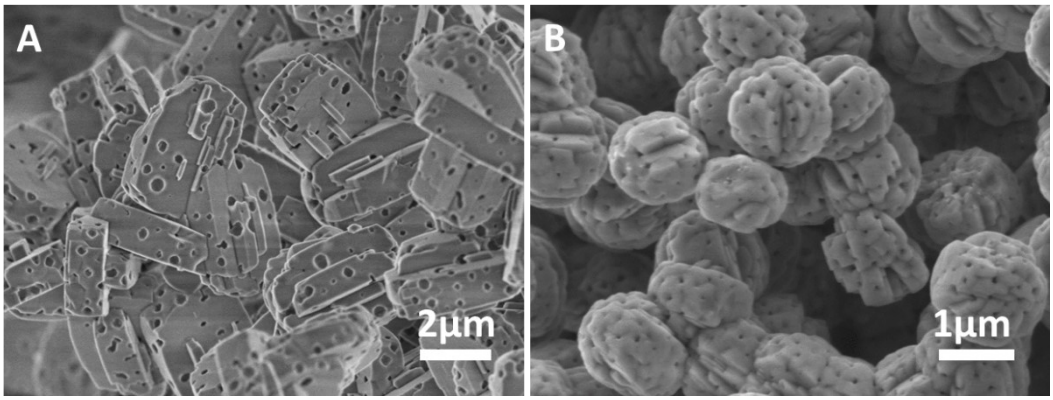

Fig. S9 SEM images of samples (A) HM-Silicalite-1(500) and (B) HM-Silicalite-1(150)

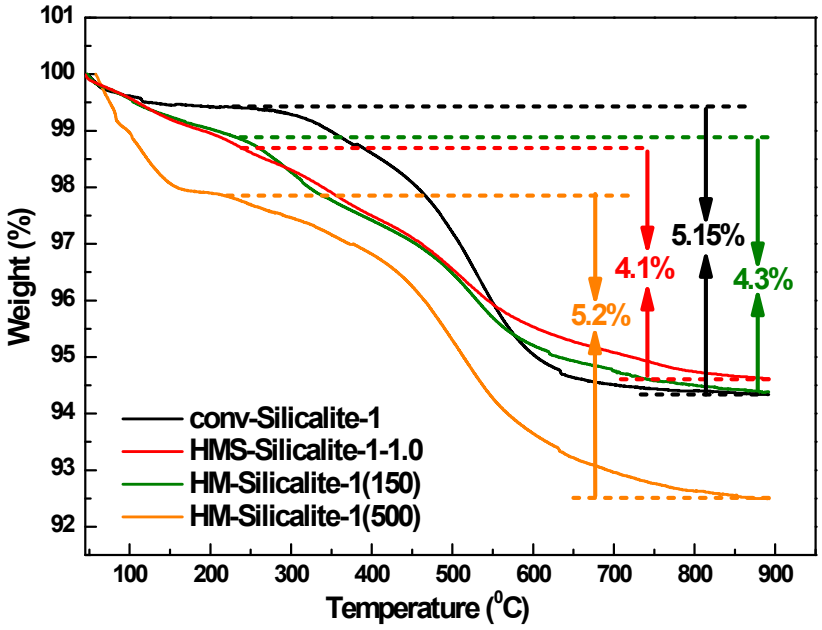

Fig. S10 TGA curves of Silicalite-1 samples after the vapor-phase Beckmann rearrangement of cyclohexanone oxime for 57 h