

Supporting information

**Insight into the dissolution-crystallization strategy towards  
macro/meso/microporous Silicalite-1 zeolites and their  
performance in the Beckmann rearrangement of  
cyclohexanone oxime**

Yanfei Zhang<sup>abc</sup>, Lanjian Xu<sup>ab</sup>, Jie Zhang<sup>ab</sup>, Peidong Li<sup>abc</sup>, Yangyang Yuan<sup>a</sup>, Hongchen Guo<sup>c</sup>,  
Xiaomin Zhang<sup>a\*</sup> and Lei Xu<sup>a\*</sup>

*<sup>a</sup> National Laboratory for Clean Energy, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023, People's  
Republic of China; Email: xulei808@dicp.ac.cn, zhangxm@dicp.ac.cn; Fax: +86 411 82463032; Tel: +86 411 82463032*

*<sup>b</sup> University of Chinese Academy of Sciences, Beijing 100049, People's Republic of China.*

*<sup>c</sup> State Key Laboratory of Fine Chemicals & School of Chemical Engineering, Dalian University of Technology, Dalian 116023, People's  
Republic of China.*

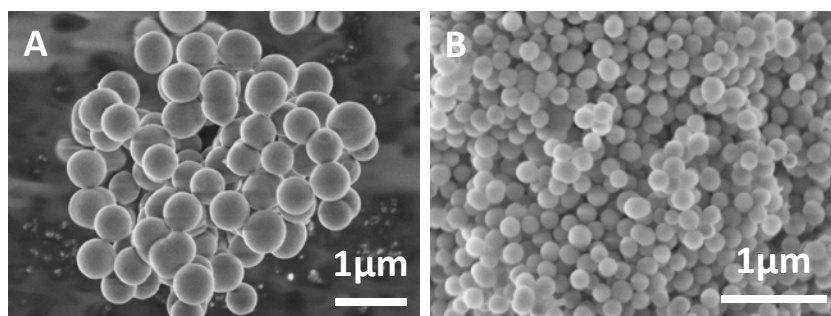


Fig. S1 SEM images of MSS-500 (A) and MSS-150 (B)

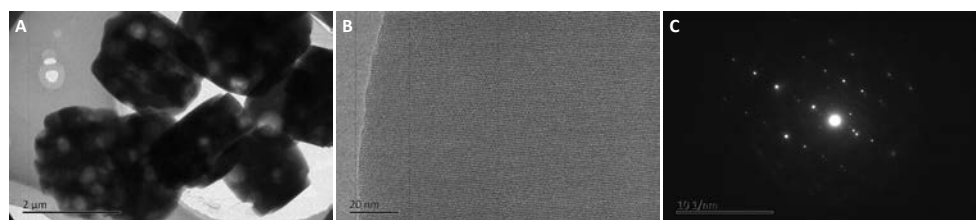


Fig. S2 Low-magnification TEM image (A), HRTEM image (B) and SAED pattern (C) of HM-Silicalite-1(500)

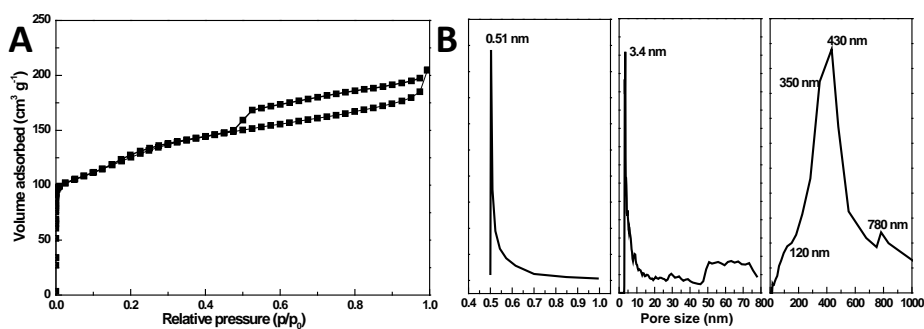


Fig. S3 N<sub>2</sub> adsorption-desorption isotherms (A) and pore size distributions (B) of the hierarchical sample HM-Silicalite-1(500) calculated by HK method, NLDFT method and Hg intrusion porosimetry, respectively

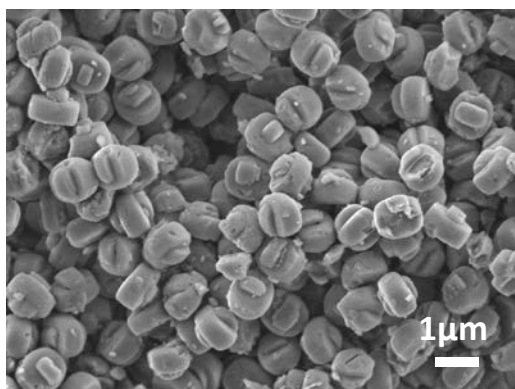


Fig. S4 SEM image of the conventional microporous sample conv-Silicalite-1

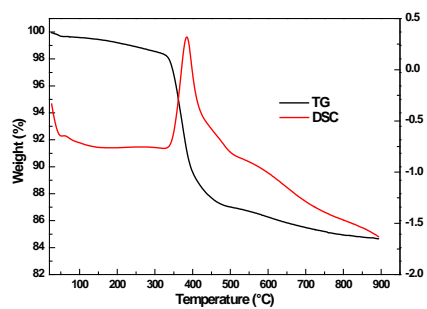


Fig. S5 TGA and DSC curves of as-prepared hierarchical sample HM-Silicalite-1(500).

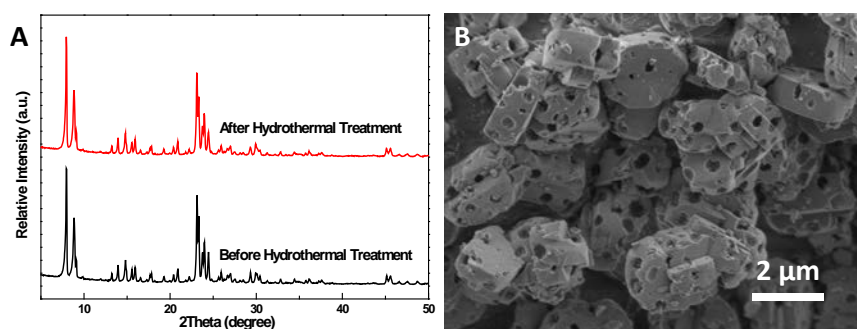


Fig. S6 Hydrothermal stability tests: (A) XRD pattern of HM-Silicalite-1 before and after hydrothermal treatment; (B) SEM image of HM-Silicalite-1 after hydrothermal treatment.

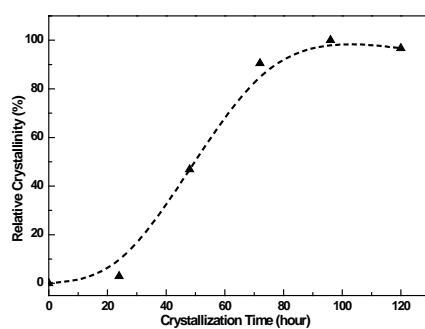


Fig. S7 Variation of relative crystallinity (RC) of HM-Silicalite-1(500) with the extension of crystallization time.

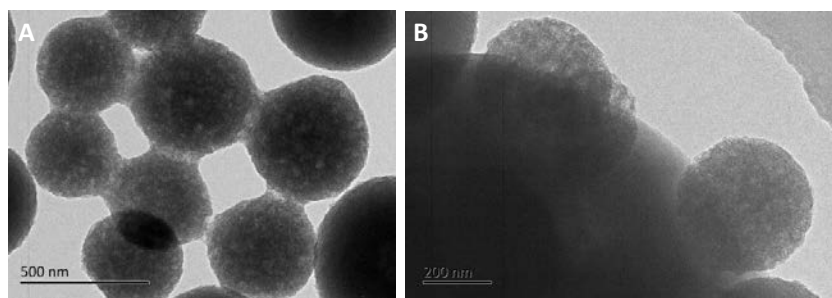


Fig. S8 TEM images of sample HM-Silicalite-1(500) crystallized at 48 h.

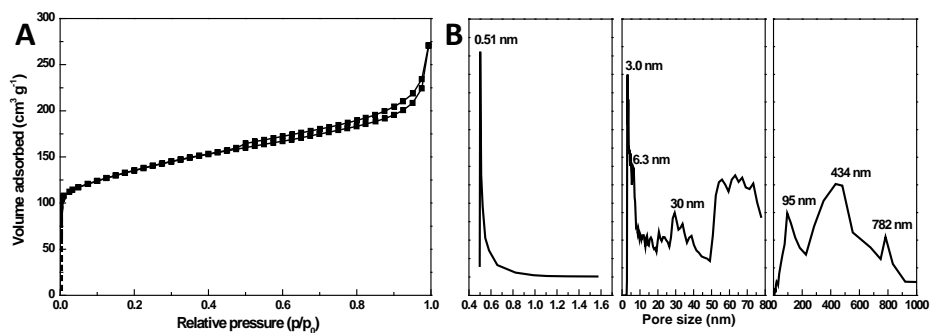


Fig. S9 N<sub>2</sub> adsorption-desorption isotherms (A) and pore size distributions (B) of hierarchical sample HM-Silicalite-1(150) calculated by HK method, NLDFT method and Hg intrusion porosimetry, respectively.

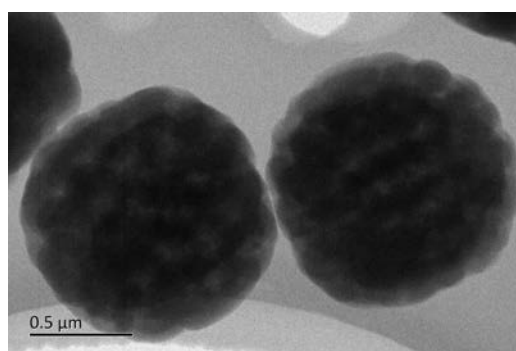


Fig. S10 Low-magnification TEM image of HM-Silicalite-1(150).

**Table S1** Synthesis conditions of the Silicalite-1 samples

Sample	Starting gel composition		Crystallization temperature/K
	TPAOH/SiO <sub>2</sub>	H <sub>2</sub> O/SiO <sub>2</sub>	
HM-Silicalite-1(500)	0.17	0.9	383
HM-S1	0.07	0.9	383
HM-S2	0.25	0.9	383
HM-S3	0.17	0.9	403
HM-S4	0.17	0.9	423
HM-S5	0.17	0.4	383
HM-S6	0.17	1.3	383
HM-S7	0.17	1.8	383
HM-Silicalite-1(150)	0.25	1.8	383