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Silicalite-1 分子筛氢键诱导晶化机制的固体核磁共振研究

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Hydrogen-Bond Induced Crystallization of Silicalite-1 Zeolite as Revealed by Solid-State NMR Spectroscopy

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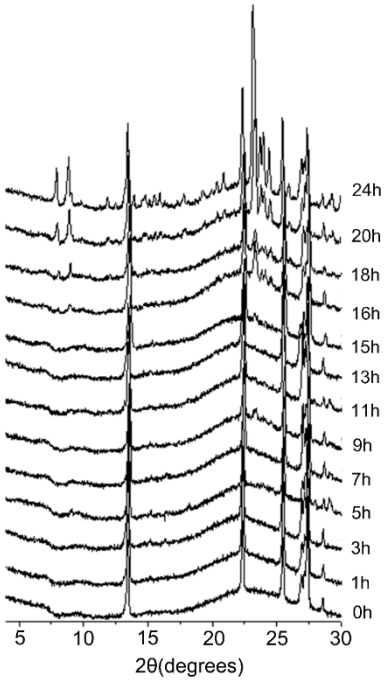

Fig. S1 XRD patterns of as-synthesized samples heated at 180 °C for different time.

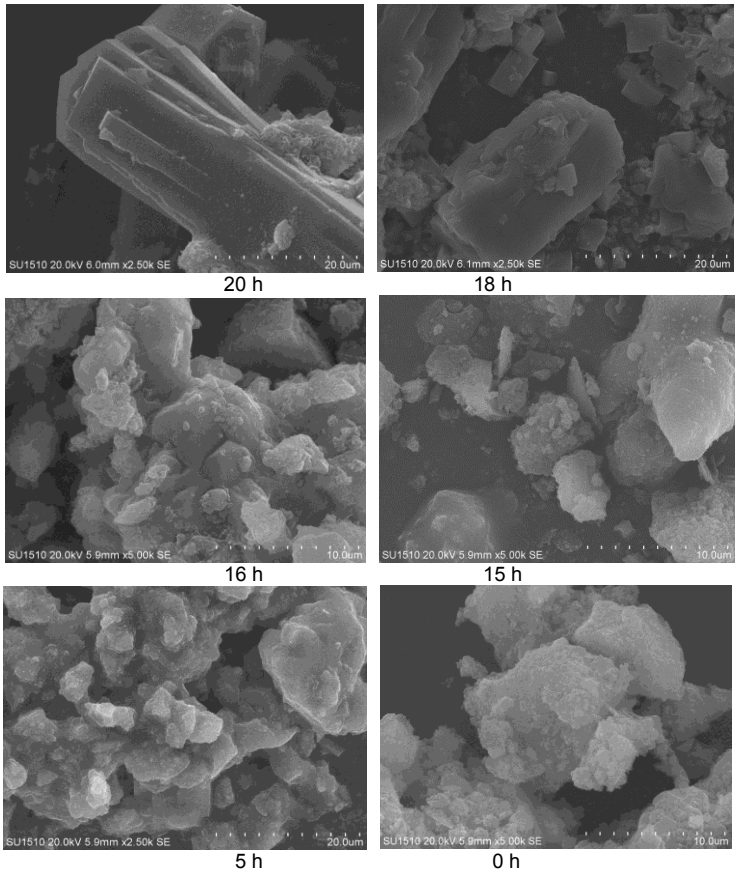

Fig. S2 SEM images of as-synthesized samples heated at 180 °C for different time.

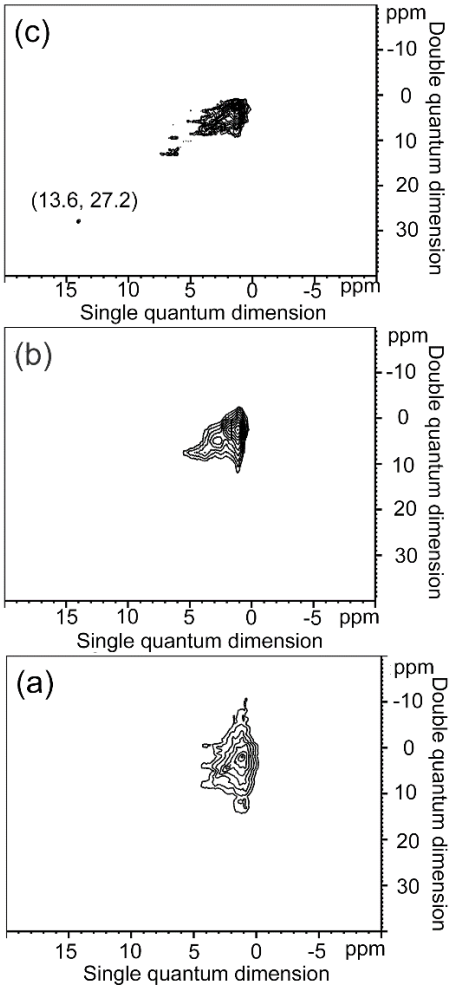

Fig. S3 ^{1}H DQ-SQ (right) NMR spectra of as-synthesized samples heated at 180 °C for (a) 3 h, (b) 7 h and (c) 11 h.

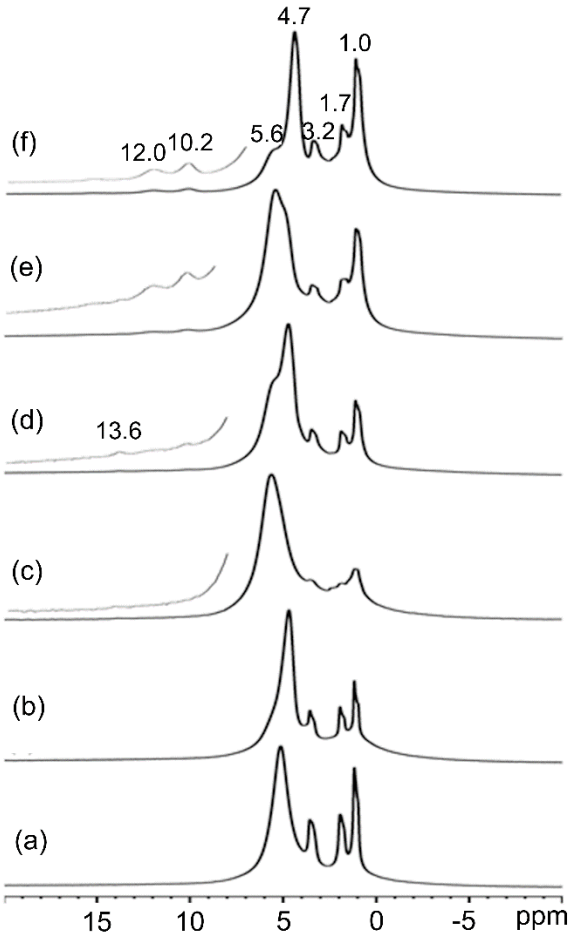

Fig. S4 ^{1}H MAS NMR spectra of as-synthesized samples acquired at different heating time: (a) 0 h, (b) 5 h, (c) 15 h, (d) 16 h, (e) 18 h and (f) 20 h (The left above part of the spectrum (6–20 ppm) is enlarged by 20 times). The signal at 13.6 ppm is from the lamellar $SiO^{-}\cdots H-OSi$ hydrogen bonds, which appears earlier than the signals at 12.0 and 10.2 ppm from in-cage $SiO^{-}\cdots H-OSi$ hydrogen bonds. The signals at 4.7 and 5.6 ppm are due to H_2O. The signals of methyl, methylene and methylene adjacent to the nitrogen groups of TPA^{+} cations are at 1.0, 1.7 and 3.2 ppm, respectively.

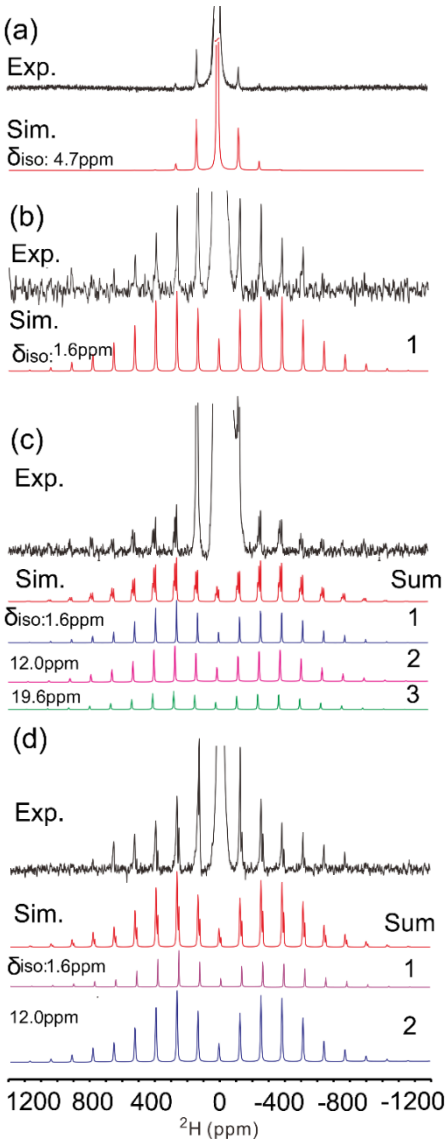

Fig. S5 Experimental (Exp.) and simulated (Sim.) ^{2}H MAS NMR spectra of as-synthesized samples heated at 180 °C for different time: (a) 15 h, (b) 16 h, (c) 18 h, (d) 20 h. For simplification, the signals of water molecules hydrogen-bonded with terminal oxygen atoms of silicates in (b), (c) and (d) are not demonstrated again.

Table S1 ^{2}H MAS NMR parameters of as-synthesized samples with increasing heating time obtained from simulations.

Heating time	δ_{iso}/ppm	QCC/kHz	Asymmetric parameter
15 h	4.7 (±0.2)	20 (±2)	0.35 (±0.08)
16 h	4.7 (±0.2)	20 (±2)	0.35 (±0.08)
	1.6 (±0.4)	120 (±5)	0.25 (±0.05)
18 h	4.7 (±0.2)	20 (±2)	0.35 (±0.08)
	1.6 (±0.4)	120 (±5)	0.10 (±0.05)
	12.0 (±0.4)	120 (±5)	0.25 (±0.05)
	19.6 (±0.4)	120 (±5)	0.25 (±0.05)
20 h	4.7 (±0.2)	20 (±2)	0.35 (±0.08)
	1.6 (±0.4)	120 (±5)	0.05 (±0.05)
	12.0 (±0.4)	120 (±5)	0.05 (±0.05)

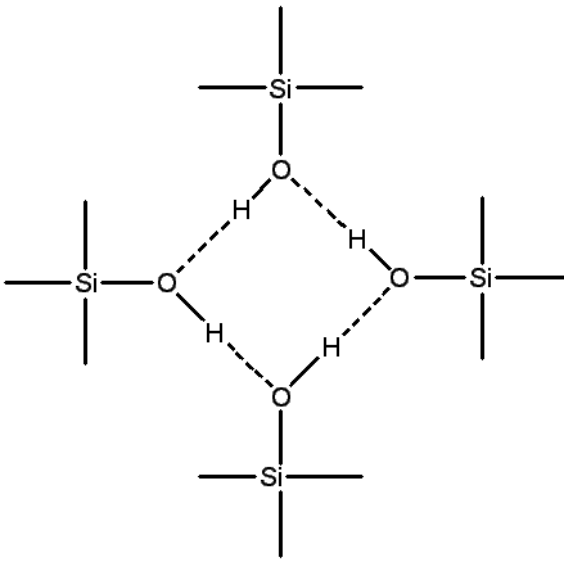

Fig. S6 Schematic of the SiOH nest.

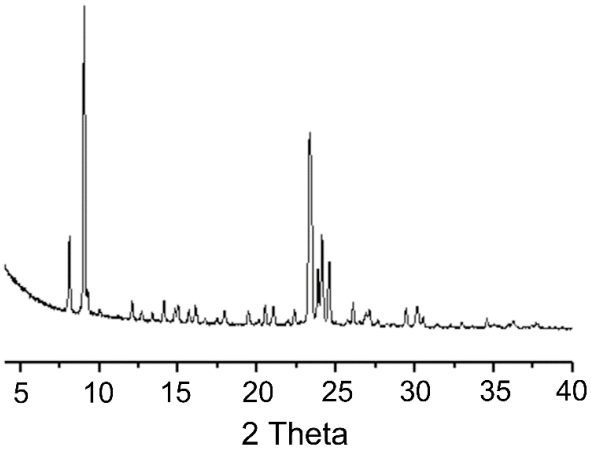

Fig. S7 XRD pattern of as-synthesized Si-ZSM-5 prepared at 175 °C under hydrothermal condition.

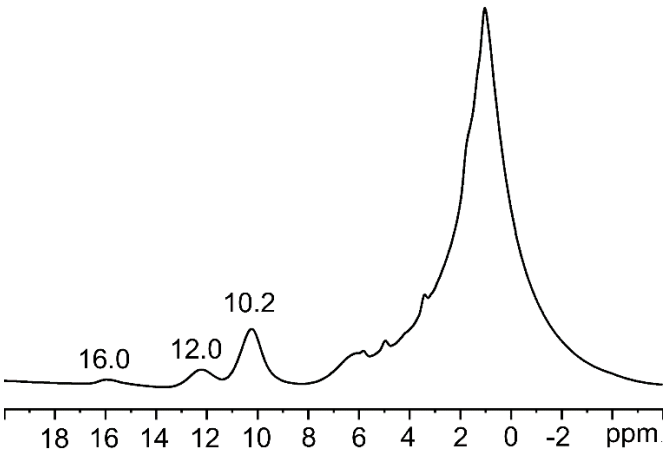

Fig. S8 ^{1}H MAS NMR spectrum of as-synthesized Si-ZSM-5 at 175 °C under hydrothermal condition.