Electronic Supplementary Information

Green Route for Synthesizing Pure Silica Zeolites with Six-Membered Rings

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Experimental Section

Materials

Sodium metasilicate nonahydrate ($Na_2SiO_3 \cdot 9H_2O$, 98%, Sinopharm Chemical Reagent Co., Ltd.), ethanol (99.8%, Sinopharm Chemical Reagent Co., Ltd.), solid silica gel (98%, Qingdao Haiyang Chemical Reagent Co., Ltd.), were used without further purification. The deionized water was made in our laboratory.

Syntheses of samples

Synthesis of pure silica zeolite with SOD structure

In a typical run for the synthesis of pure silica zeolite with SOD structure, 0.3 g of $Na_2SiO_3 \cdot 9H_2O$, 1.56 g of solid silica gel, and 0.1 g of SOD seeds were mixed, followed by addition of 1.9 g of ethanol. After heating at 140 °C for 4 days in an autoclave, the product was finally crystallized, which was designated as S-SOD. Notably, the obtained S-SOD product could also be used as seed crystals for synthesizing pure silica S-SOD-2nd zeolite.

Synthesis of pure silica zeolite with MTN structure

In a typical run for the synthesis of pure silica zeolite with MTN structure, 0.3 g of $Na_2SiO_3 \cdot 9H_2O$, 1.56 g of solid silica gel, and 0.1 g of MTN seeds were mixed, followed by addition of 1.9 g of ethanol. After heating at 180 °C for 2 days in an autoclave, the product was fully crystallized, which was designated as S-MTN.

Synthesis of pure silica zeolite with NON structure

In a typical run for the synthesis of pure silica zeolite with NON structure, 0.65 g of $Na_2SiO_3 \cdot 9H_2O$, 1.54 g of solid silica gel, and 0.1 g of NON seeds were mixed, followed by addition of 1.9 g of ethanol. After heating at 180 °C for 1 day in an autoclave, the product was fully crystallized, which was designated as S-NON.

Characterization

X-ray powder diffraction (XRD) patterns were measured with a Rigaku Ultimate VI X-ray diffractometer (40 kV, 40 mA) using CuKα (λ=1.5406 Å) radiation. TG-DTA analysis was carried out with a NETZSCH STA 449C in air at a heating rate of 10 °C/min from room temperature to 800 °C. The Fourier Transform-Infrared Spectroscopy (FT-IR) spectra were recorded using a Bruker 66V FT-IR spectrometer. Scanning electron microscopy (SEM) experiments were performed on Hitachi SU-1510 electron microscopes. ^{29}Si, ^{13}C, and ^{1}H MAS NMR spectra were recorded on a Bruker AVANCEIII 500WB spectrometer.

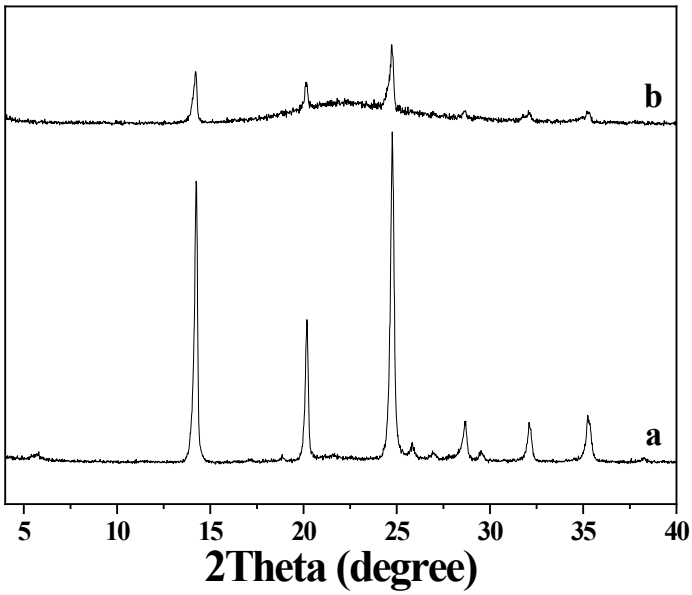

Figure S1 XRD patterns of the products synthesized from the starting mixture with molar ratio of 1.0 SiO_2/0.04 Na_2O/6% seeds with C_2H_5OH/SiO_2 ratios of (a) 0.5, and (b) 3, respectively.

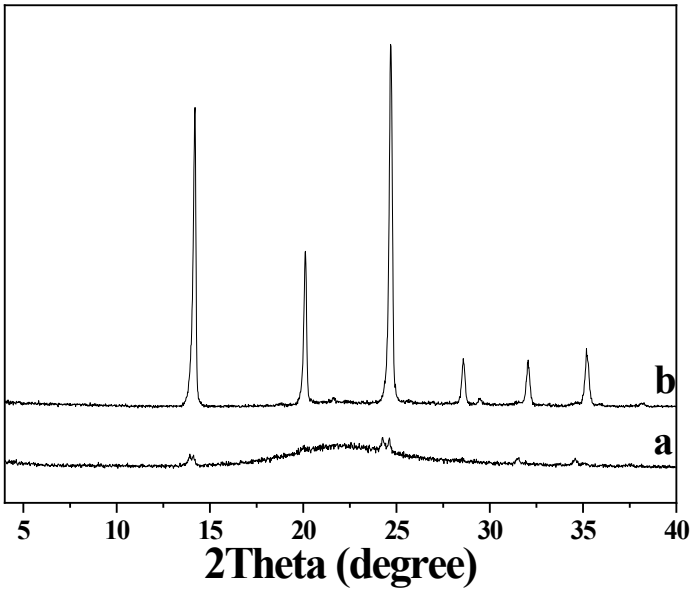

Figure S2 XRD patterns of the products synthesized from the starting mixture with molar ratio of 1.0 SiO_2/1.5 C_2H_5OH/6% seeds with Na_2O/SiO_2 ratios of (a) 0.03, and (b) 0.05, respectively.

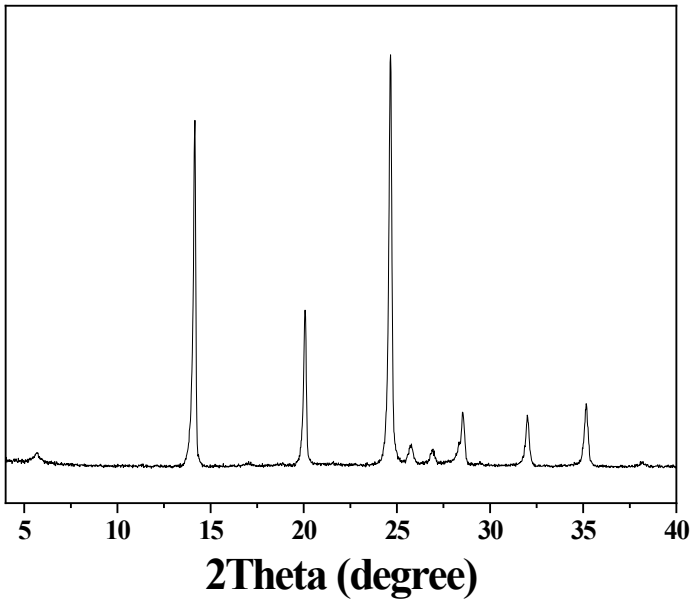

Figure S3 XRD pattern of the product synthesized from the starting mixture with molar ratio of 1.0 SiO_2/0.04 Na_2O/1.5 C_2H_5OH/3 H_2O/6% seeds.

Figure S4 Photographs of (a) the initial mixtures before crystallization and (b) the product after crystallization, respectively.

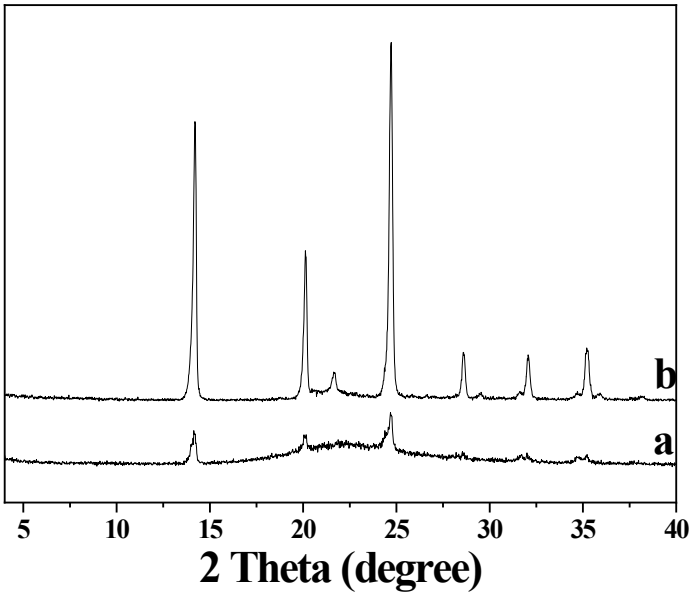

Figure S5 XRD patterns of the products synthesized from the starting mixture with molar ratio of 1.0 SiO_2/0.04 Na_2O/1.5 C_2H_5OH/6% seeds at (a) 120 °C, and (b) 160 °C, respectively.

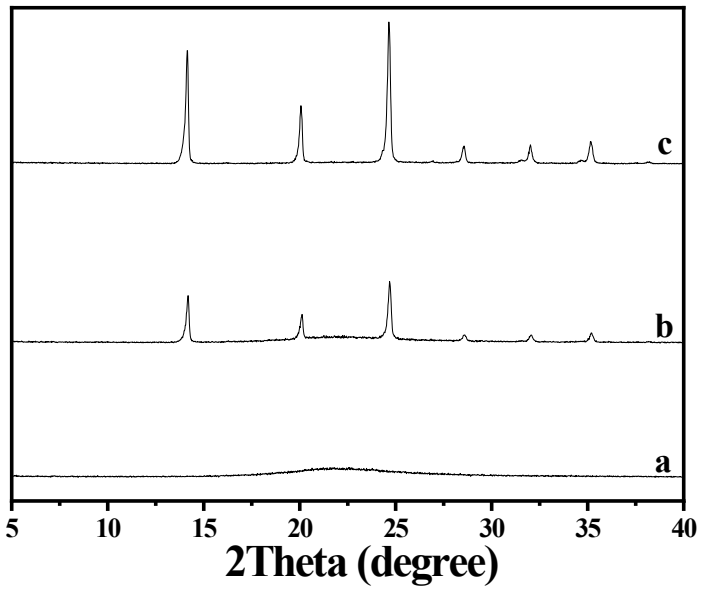

Figure S6 XRD patterns of the products synthesized from the starting mixture with molar ratio of 1.0 SiO_2/0.04 Na_2O/1.5 C_2H_5OH with mass ratios of zeolite seeds to the silica source at (a) 0%, (b) 2.5%, and (c) 10%, respectively.

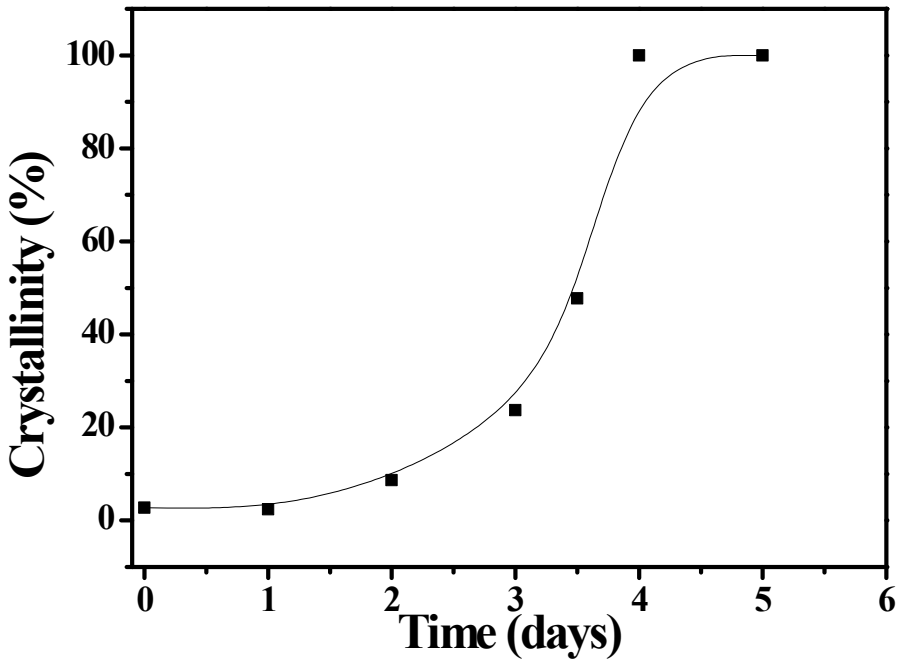

Figure S7 The dependence of pure silica SOD zeolite crystallinity over crystallization time. The S-SOD crystallinity is based on its peak intensity at 24.8° in XRD pattern, and fully crystallized S-SOD crystallized at 140 °C for 4 d is designated as 100 % crystallinity.

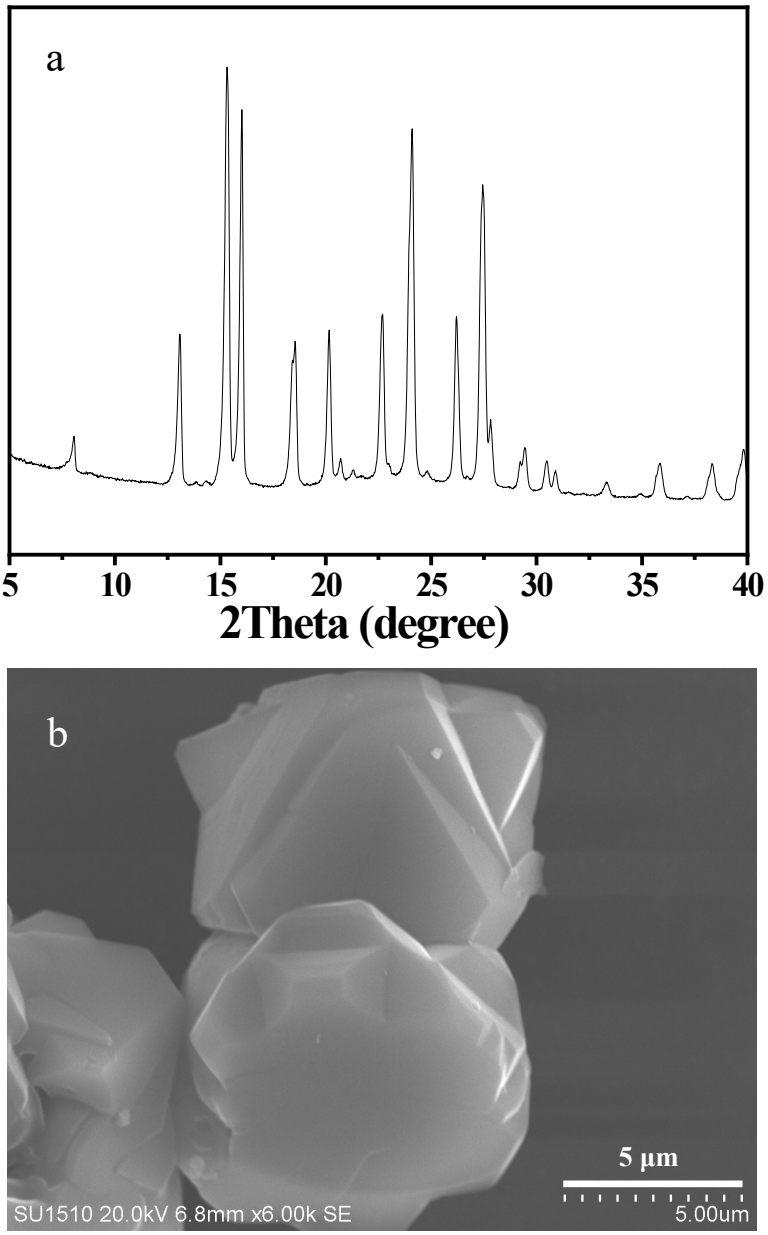

Figure S8 (a) XRD pattern and (b) SEM image of pure silica MTN zeolite.

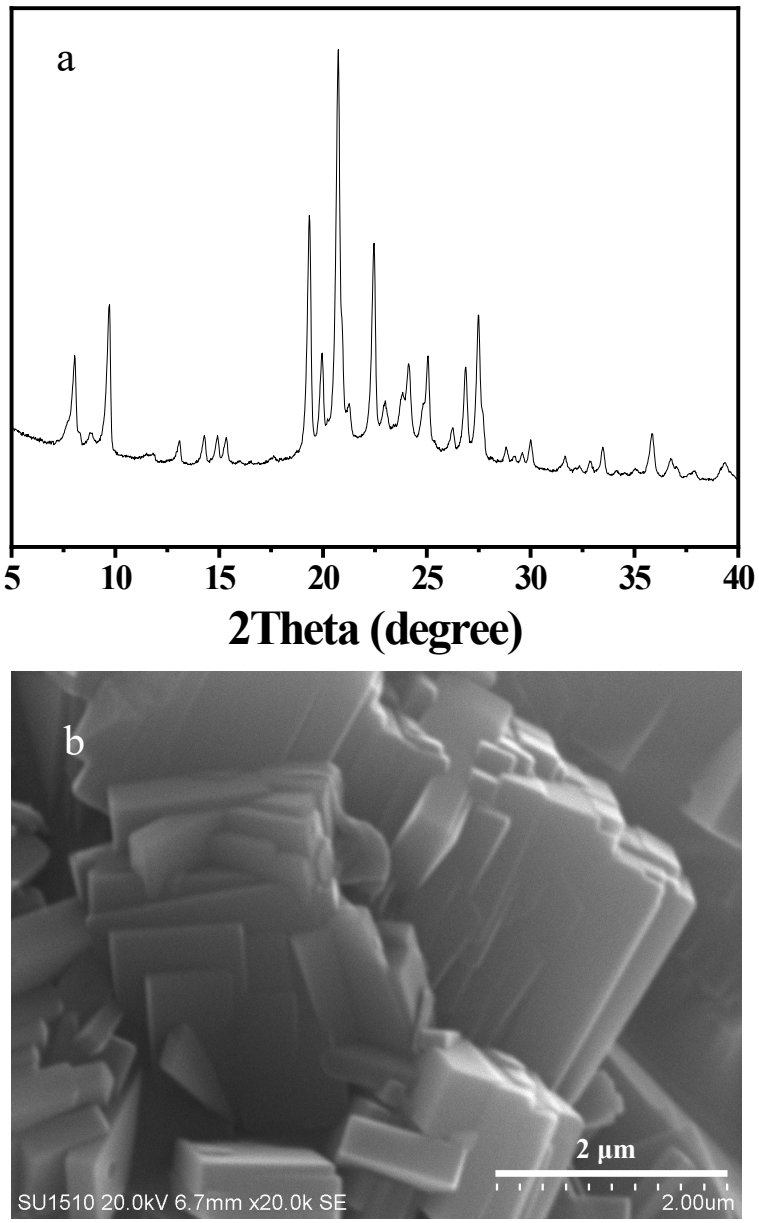

Figure S9 (a) XRD pattern and (b) SEM image of pure silica NON zeolite.

Table S1. ^{29}Si NMR data of S-SOD samples synthesized for 0-4 days

Time (days)	Si(4Si) (ppm)	Si(4Si) (ppm)	Si(3Si) (ppm)
0		-111.0	-100.6
1		-111.2	-100.3
2	-116.3	-111.5	-102.6
3.5	-116.7	-111.6	-100.3
4	-116.5	-111.1	-100.5

Table S2. Structural information on S-SOD samples from ^{29}Si NMR analysis

Time (days)	Si(4Si) (%)	Si(4Si) (%)	Si(3Si) (%)
0		81.2	18.8
1		84.3	15.7
2	2.0	68.1	29.9
3.5	9.9	74.7	15.4
4	64.7	26.1	9.2