Electronic Supplementary Information

Hydrothermal synthesis of high silica zeolite Y using tetraethylammonium hydroxide as structure-directing agents

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Synthesis

TEA-Y samples. The process of synthesizing high silica Y zeolite with TEAOH as SDAs was as follows: As a typical run of the sample TEA-1, sodium metaaluminate (Al_2O_3 = 48%, Na_2O = 40%, Sinopharm Chemical Reagent Corp.) and sodium hydroxide (98%, Tianjin Kemiou Chemical Reagent Corp.) were mixed with tetraethylammonium hydroxide (35% aq. Solution, Shanghai Aladdin Chemical Reagent Corp.) in deionized water and stirred to a clear solution. Then, a given amount of silica gel (29.8% aq. Solution, Qingdao Ocean Chemical Plant) was slowly added into the solution. Composition of the starting gels is 1.3 Na_2O/1.5 TEA_2O/10 SiO_2/1 Al_2O_3/90 H_2O. This suspension was aged with agitation at room temperature for 12 h, and then poured into a stainless steel autoclave, and placed in an oven statically at 120 ºC for 14 days. After crystallization, the as-synthesized products were separated by filtration, washed with deionized water until pH < 8, and then dried at 100 ºC for further characterization.

Na-Y sample. The process of synthesizing sample Na-Y was as follows: sodium metaaluminate and sodium hydroxide were added in deionized water and stirred to a clear solution. Then, a given amount of silica gel was slowly added into the solution. The composition of the starting gels is 3.2 Na_2O/10 SiO_2/1 Al_2O_3/90 H_2O. This suspension was aged with agitation at room temperature for 12 h, and then poured into a stainless steel autoclave, and placed in an oven statically at 120 ºC for 14 days. After crystallization, the as-synthesized products were separated by filtration, washed with deionized water until pH < 8, and then dried at 100 ºC for further characterization.

H-form TEA-Y zeolites. H-form zeolites were prepared by ion-exchange of NH_4NO_3 solution, followed by calcination. In a typical run, 2 g calcinated zeolite was ion-exchanged with 50 mL of NH_4NO_3 solution (1.5 mol/L) at 80 °C for 5 h. After washing with deionized water and drying at 100 °C for 12 h, the ion-exchanged zeolite was calcined at 500 °C for 4 h. This process was repeated for two times.

Hydrothermal treatment. Hydrothermal treatment of the H-form zeolites was performed in a tube furnace. The H-form zeolites were placed in the tube furnace and were heated in 100 % steaming vapor at temperature of 750 °C. The samples were designated as zeolites-hydro.

Characterization

The X-ray diffraction (XRD) patterns were collected on a PANalytical X'Pert PRO X-ray diffractometer using the Cu-Kα radiation (λ = 1.54059 Å), operating at 40 kV and 40 mA. The step size was 0.02 °, and the scanning speed was 12 °/min. The relative crystallinity was estimated by the reflection intensities of the peaks (1 1 1), (2 2 0), and (3 3 1) of the samples. The crystal size and morphology were observed with a Hitachi SU8020 scanning electron microscopy. N_2 adsorption-desorption isotherms of the samples were measured at 77 K on a Micromeritics ASAP 2020 system. All the solid state NMR experiments were performed on a Bruker AvanceIII 600 spectrometer equipped with a 14.1 T wide-bore magnet. TG-DSC analysis was carried out on TA Q-600 analyzer from room temperature to 1200 ºC with a heating rate of 10 ºC/min in an air flow of 100 ml/min. the chemical composition of the samples was calculated with a Philips Magix-601 X-ray fluorescence (XRF) spectrometer. Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES) analysis was measured with PerkinElmer 7300DV.

Supporting Figures

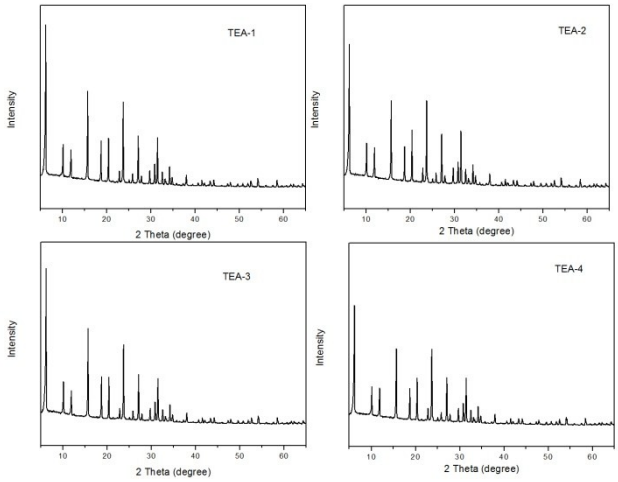

Fig. S1 XRD patterns of the as-synthesized samples TEA-1, TEA-2, TEA-3, and TEA-4.

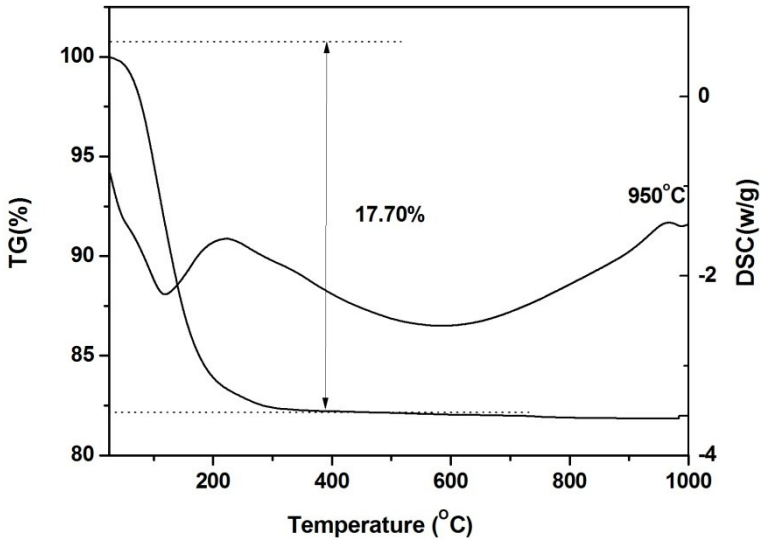

Fig. S2 TG-DSC curves of the sample Na-Y.

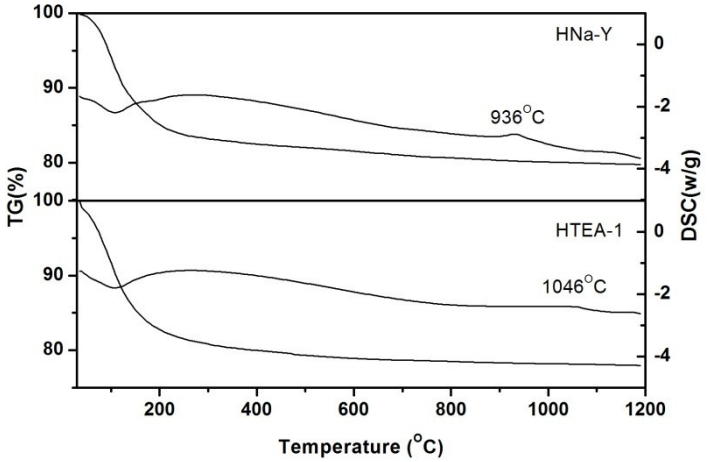

Fig. S3 TG-DSC curves of the H-form zeolites Na-Y and TEA-1.

Table S1. Products synthesized under various conditions from the gel. Composition of the starting gels is x Na_2O/y TEA_2O/10 SiO_2/1 Al_2O_3/90 H_2O.

Run	x	y	Cryst. Time (d)	Cryst. temp. (℃)	Products
1	2.2	1.0	14	120	FAU+GIS
2	1.7	1.5	14	120	FAU+GIS
3	1.8	1.4	14	120	FAU+GIS
4	1.0	1.8	21	120	FAU+BEA
5	1.1	1.7	21	120	FAU+BEA
6	1.2	1.6	21	120	FAU+BEA
7	1.2	1.8	14	120	FAU+BEA
8	1.3	1.5	14	120	FAU
9	1.5	1.5	14	120	FAU
10	1.5	1.3	14	120	FAU
11	1.8	1.2	12	120	FAU
12	1.8	1.0	12	120	FAU
13	2.0	1.0	8	120	FAU
14	2.1	0.7	8	120	FAU
15	2.1	0.9	4	120	FAU
16	2.3	0.7	4	120	FAU+MOR
17	2.3	0.5	4	120	FAU+MOR
18	2.5	0.5	4	120	FAU+MOR
19	2.5	0.3	8	120	MOR
20	1.8	0.8	14	120	Amor
21	1.2	1.4	14	120	Amor
22	1.4	1.2	14	120	Amor
23	1.6	1.0	14	120	Amor