A Multiple-Assembly/One-Pot-Crystallization Strategy for a Relatively More Eco-friendly Synthesis of Hydrothermally Stable Mesoporous Aluminosilicates

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Supplementary materials

Experimental section

1. Materials

Triblock copolymer P123 ($EO_{20}PO_{70}EO_{20}$, Mw=5800) was purchased from Aldrich. Water glass (containing 28.3 % SiO_2 and 8.8 % Na_2O), sodium hydroxide (NaOH), sulfuric acid (H_2SO_4) and aluminium sulfate octadecahydrate ($Al(SO_4)_3 \cdot 18H_2O$) were obtained from Tianjin Guangfu company. Deionized water was obtained from Beijing University of Chemical Technology.

2. Experimental

2.1. Preparation of zeolite Y precursors:

The Y precursors were prepared according to literature [6].

2.2. Multiple assembly

(1) 300 g zeolite Y precursors and 6 M H_2SO_4 were simultaneously added into the

P123 solution (40 g P123 disolved in 1500 mL water) at pH 1.5. The mixture gel was assembled at 30 °C for 20 h to obtain “assembly product” and “assembly mother liquor”. Assembly mother liquor and assembly product (filter cake 1) was separated by filtration.

(2) Prior to the second assembly cycle, 15 g P123 and 240 g Y precursors, were added to the assembly mother liquor obtained in the last step. H_2SO_4 was added slowly under stirring until pH of the mixture was about 1.5. After the same assembly process with that of step (1), assembly mother liquor and assembly product (filter cake 2) was separated by filtration.

(3) The third, fourth, and fifth assembly cycles were repeated as the procedure described in step (2). The assembly products obtained from the third, fourth and fifth cycles will be denoted as “filter cake 3”, “filter cake 4”, and “filter cake 5”.

2.3 Crystallization:

Filter cakes 1, 2, 3, 4, and 5 were mixed with the mother liquor (the recycling liquor in cycles 1-5) and the mixture was crystallized at 120 °C for 24 h in a 1 L Teflon autoclave. After filtration, washing, drying and calcination at 550 °C for 5 h, the final product was obtained and denoted as MAOC-5 (mother liquor was cycled for 5 cycles). The flow chart is shown in Fig. 1.

For comparison, MA-1, MA-2, MA-3, MA-4, and MA-5 were obtained by direct crystallization of “assembly mother liquor” and “assembly product” obtained in step (1), (2), (3), (4), and (5) respectively at 120 °C for 24 h.

3. Characterization

X-ray diffraction (XRD) patterns of the synthesized aluminosilicate were obtained with a Rigaku D/Max 2500VB2+/PC diffractometer using Cu Kα radiation. Transmission electron microscopy (TEM) images were recorded by JEM 100CX with an acceleration voltage of 200 kV. The isotherms of nitrogen were measured at the temperature of liquid nitrogen using a Micromeritics ASAP 2405N system. The pore-size distribution was calculated using the Barrett-Joyner-Halenda (BJH) model. FTIR spectra were measured with spectrometer of Nicolet 8700, and KBr was using as an internal standard sample.

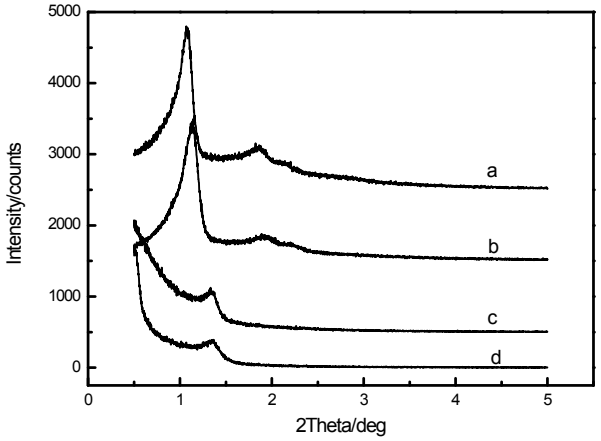

Fig. S1 XRD patterns for (a) MA-1, (b) MAOC-5, (c) HMA-1, and (d) HMAOC-5

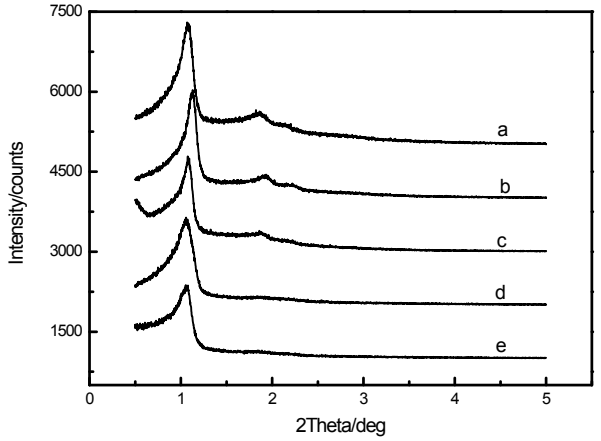

Fig. S2 XRD patterns for (a) MA-1, (b) MA-2, (c) MA-3, (d) MA-4 and (e) MA-5

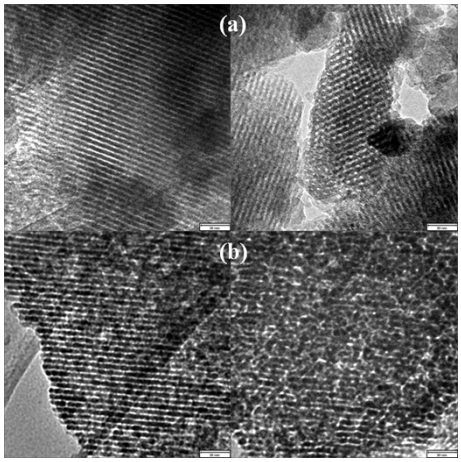

Fig. S3 The TEM images of (a) MAOC-5 and (b) HMAOC-5

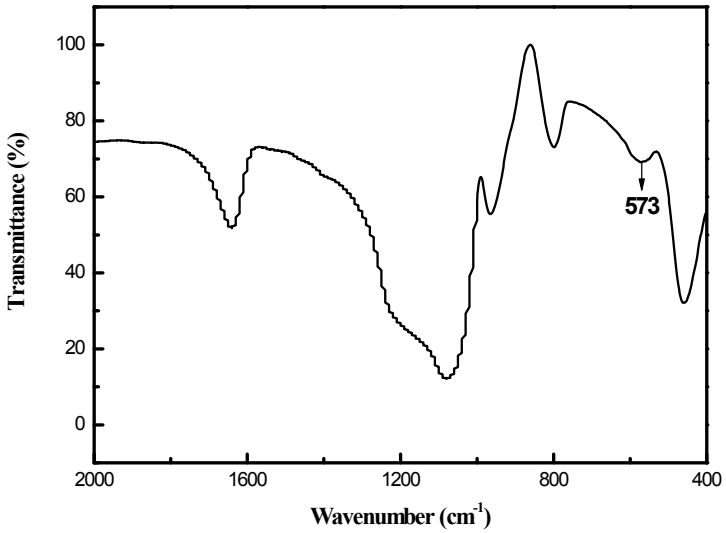

Fig. S4 FT-IR spectra of MAOC-5 sample