Supporting Information

Controlled Release of Siliceous Species for the Fabrication of Highly *b*-Oriented MFI Zeolite Films

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Materials: Stainless steel plates were immersed in hydrogen peroxide solution for 45 min, then rinsed with deionized water, and dried at 60 °C before the synthesis of zeolite films. Tetraethyl orthosilicate (TEOS, 98%) and 1,2-dihydroxybenzene (99%) were purchased from J&K Scientific Ltd.. Tetrapropylammonium hydroxide (TPAOH, 25wt. %) was supplied by Sachem Wuxi.

Preparation of MFI zeolite seeds: The synthesis solution of molar composition 0.17 TPAOH:1 TEOS:165 H_2O was prepared by slowly adding TEOS to a solution of TPAOH and water under stirring. A clear synthesis solution was obtained after stirring at room temperature for 4 h. Then the synthesis solution was directly loaded into a Teflon-lined stainless steel autoclave. The autoclave was sealed and fixed in the rotation shaft of a convectional oven. It rotated with the axis at 20 rpm in the oven at 175 °C for 130 min. After synthesis, the mixture was quenched. The sample was recovered, thoroughly washed with deionized water, and dried at 60 °C. The size of the MFI zeolite seed crystals is ca. 1 μm.

Preparation of *b*-oriented MFI zeolite films: Perfectly *b*-oriented MFI seed monolayers were prepared on film supports by rubbing MFI zeolite crystals with a finger in latex glove and then heat treated at 165 °C for 4 h. The seeded support was vertically placed in a Teflon-lined stainless steel autoclave for secondary growth. The synthesis solution was prepared by slowly adding TEOS to a solution of TPAOH and water under stirring. After the solution was clear, 1,2-dihydroxybenzene was added into the synthesis solution. The molar composition of the resulting solution was 0.2TPAOH : 1TEOS : 200H_2O : (0~0.5)$C_6H_6O_2$. The seeded growth was carried out at 165 °C for 4 h. After synthesis, the film was rinsed with deionized water and dried at 60 °C.

Characterization: The top and cross-sectional images of the zeolite films were obtained with scanning electron microscope SU-8010 (Hitachi) and JSM-6701F (JEOL). To observe the cross-sectional morphology, the edges of stainless steel supported MFI zeolite films were immersed into diluted HF aqueous solution for a few seconds. Then the sample were washed with deionized water, and dried at 60 °C. X-ray diffraction (XRD) patterns were collected on a Bruker D8 Advance diffractometer using Cu *K*α radiation. ^{29}Si-NMR spectra of the mother solutions for MFI film synthesis were recorded on a Bruker Avance Ⅲ HD 500 spectrometer operating at a resonance frequency of 99.292 MHz.

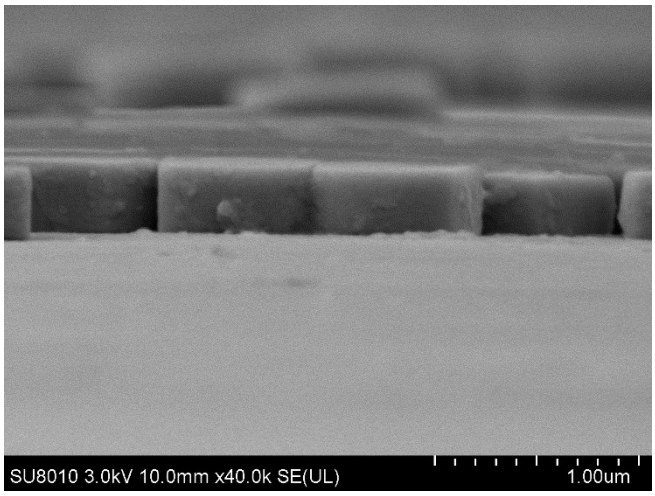

Figure S1 Cross-sectional SEM image of zeolite MFI seed layer.

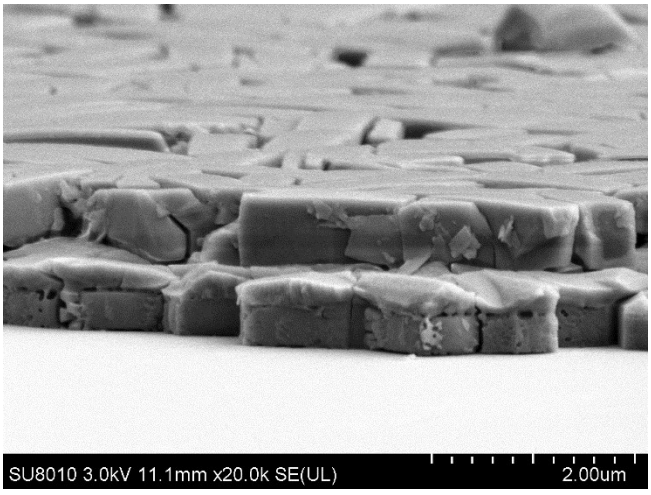

Figure S2 Cross-sectional SEM images of MFI films synthesized at 165 ^{o}C for 4 h. Synthesis solution composition: 0.2TPAOH : 1TEOS : 200H_2O.

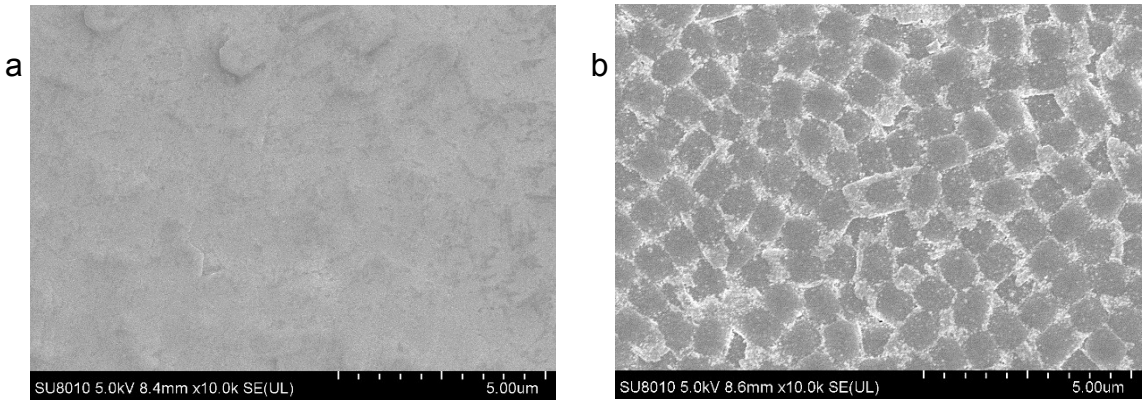

Figure S3 SEM images of MFI films synthesized at 165 °C for 4 h. Synthesis solution composition: 0.2TPAOH : 1TEOS : 200H_2O : $xC_6H_6O_2$, (a) x = 0.4, (b) x = 0.5.

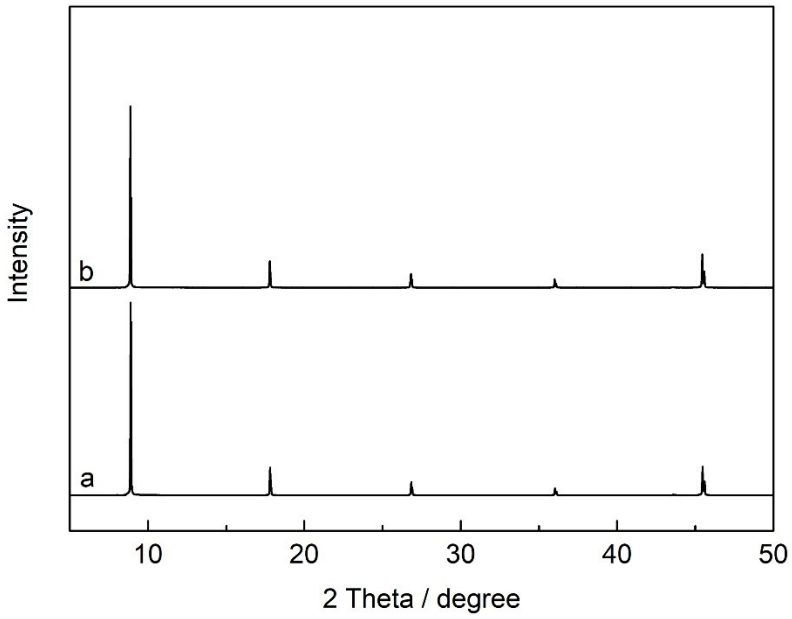

Figure S4 XRD patterns of MFI films synthesized at 165 °C for 4 h. Synthesis solution composition: 0.2TPAOH : 1TEOS : 200H_2O : $xC_6H_6O_2$, (a) x = 0.4, (b) x = 0.5.