[Supplementary Information]

Fig. S1 Electron micrographs showing the crystal ripening of nano-sized zeolite framework. (a, c) SEM, and **(b, d)** TEM images of the products precipitated after 30 h and 12 d, during hydrothermal synthesis of mesoporous MFI zeolite at 150°C. The images for 12 d show significant ripening of the nanocrystalline zeolite frameworks, compared with 30 h. As shown by the TEM images, the ripening was particularly conspicuous at the external surface.

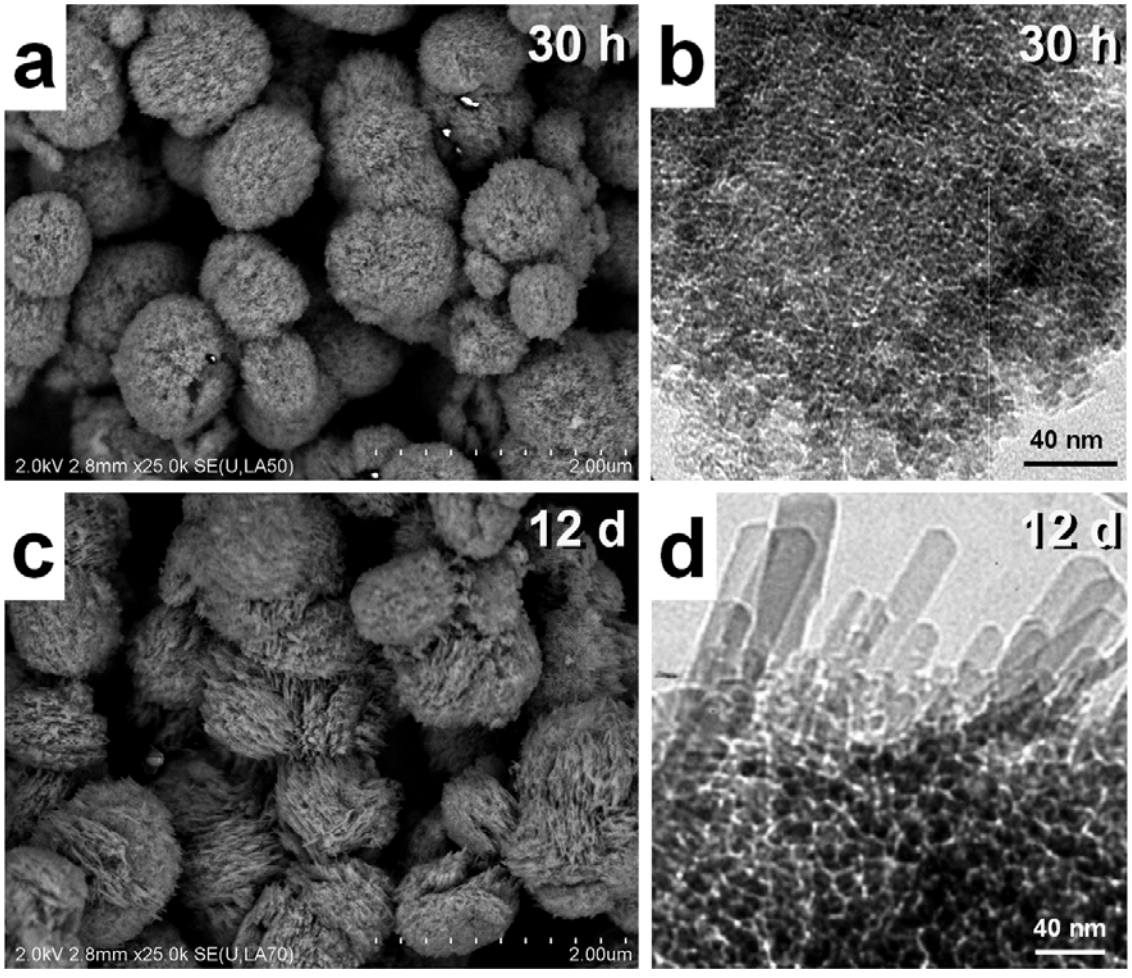

Fig. S2 Pore structure analysis of the products precipitated after various reaction times during hydrothermal synthesis of mesoporous MFI zeolite at 130°C. (a) N_2 adsorption isotherms, and **(b)** distribution of mesoporous diameters. The isotherms for 8 d and 12 d were offset vertically by 200 and 400 mL g^{-1}. Distributions of mesopore diameters were calculated via BJH algorithm using the adsorption branch[33]. The distribution of mesoporous diameters for 8 d and 12 d were offset vertically by 2 and 4 mL $g^{-1}nm^{-1}$, respectively. The mesopore diameters did not significantly change until 12 d, due to the slow crystal ripening process compared with 150°C.

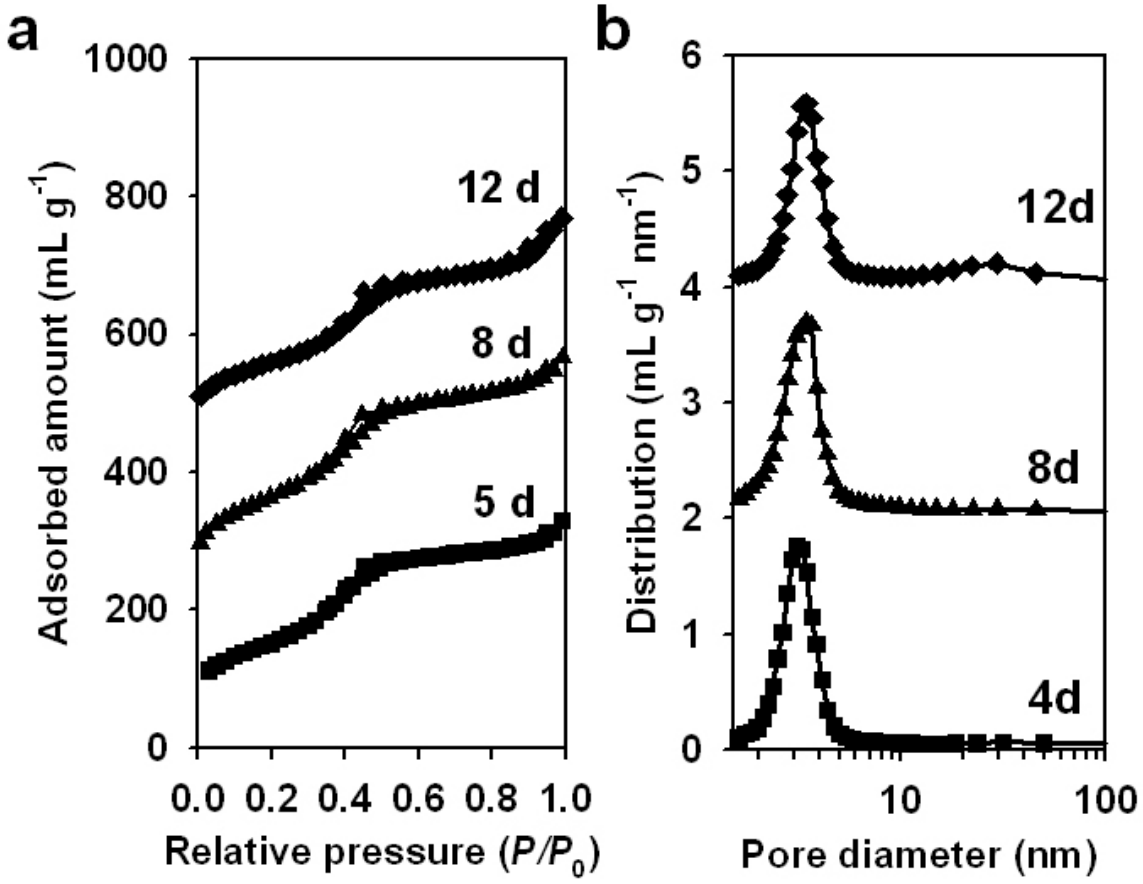

Fig. S3 Electron micrographs showing the uniform formation of mesoporous MFI zeolites. (a, c) SEM, and **(b, d)** TEM images of the mesoporous MFI zeolites precipitated after 5 d hydrothermal synthesis at 130°C. The images show that the hydrothermal reaction resulted in the complete transformation of amorphous gel into zeolite (globular morphology with rugged surface). Bulk (not mesoporous) zeolite crystals were not found.

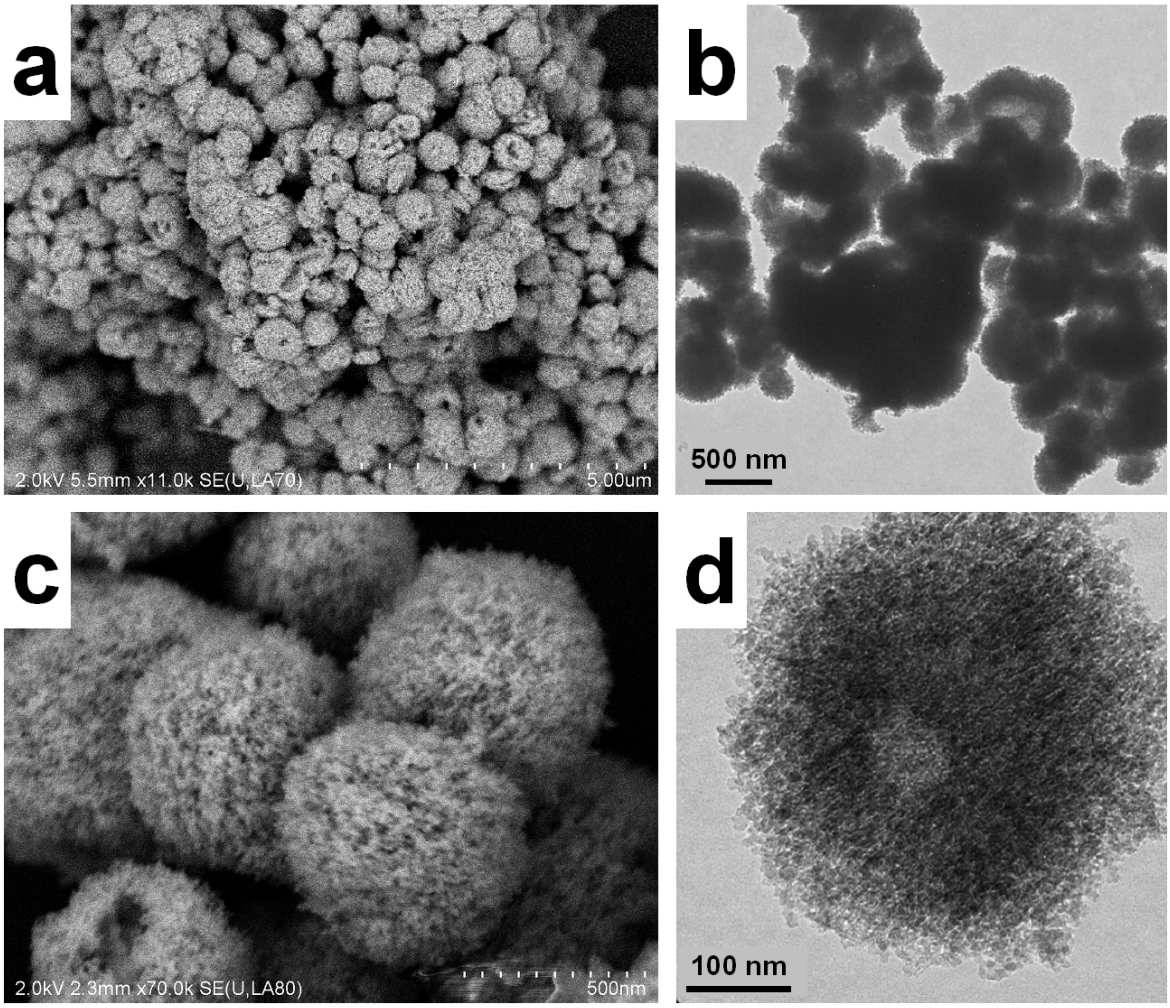

Fig. S4 Solid-state MAS ^{27}Al NMR spectrum of mesoporous MFI zeolite. The measurements were recorded on a solid-state Bruker AM-300 NMR spectrometer and using a Bruker 7 mm MAS probe. A typical spinning rate for MAS experiments is 4 kHz. ^{27}Al NMR spectrum was acquired with 0.5 s repetition time, 3 μs delay time and 2000 scans. ^{27}Al was referenced to an $Al(NO_3)_3$ aqueous solution.

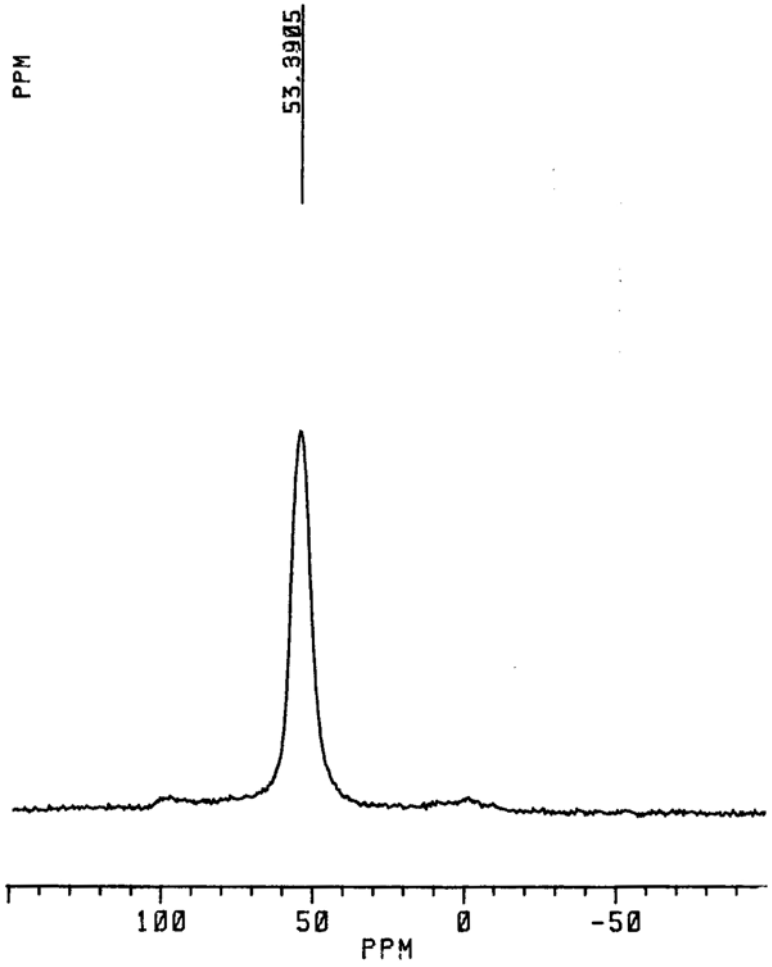

Fig. S5 Ammonia TPD profiles of the various aluminosilicate materials (Si/Al=20) presented in Table 1. For the measurement, 50 mg powder samples were introduced into a quartz reactor and degassed under vacuum at 550°C. After cooling down to room temperature, ammonia gas was adsorbed on the samples for 1 h, followed by evacuation at 120°C for removing free and weakly adsorbed ammonia. TPD profiles were then measured with evacuation at the temperature gradient of 10°C min^{-1}, using a quadruple mass detector.