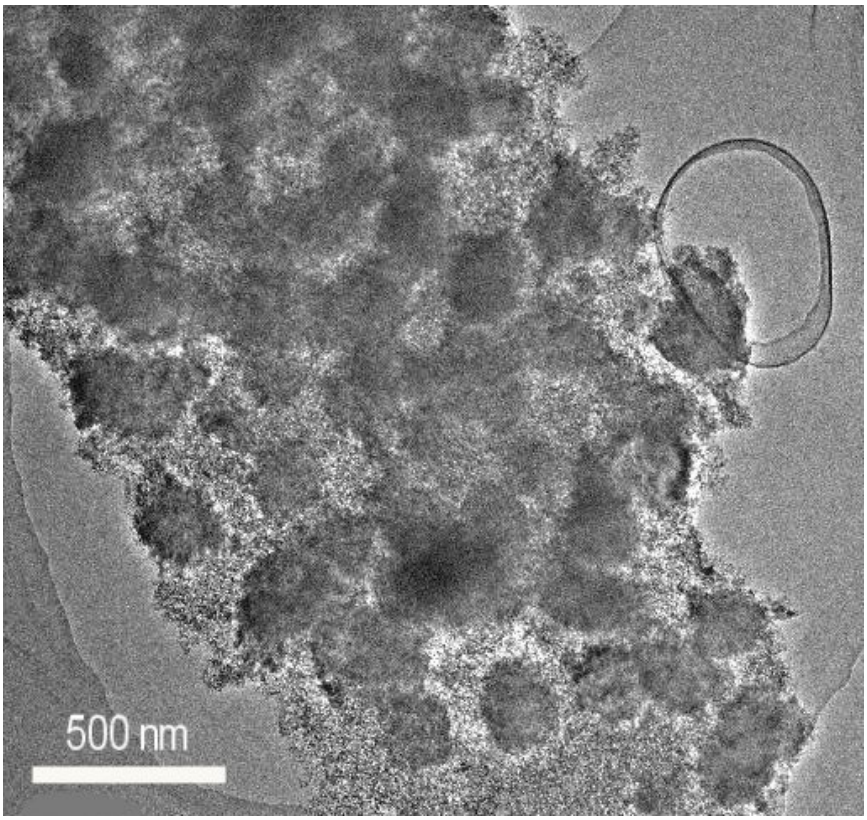

Fig. S1 TEM image of sample 10 h. The crystals have larger sizes than in the 3 h sample.

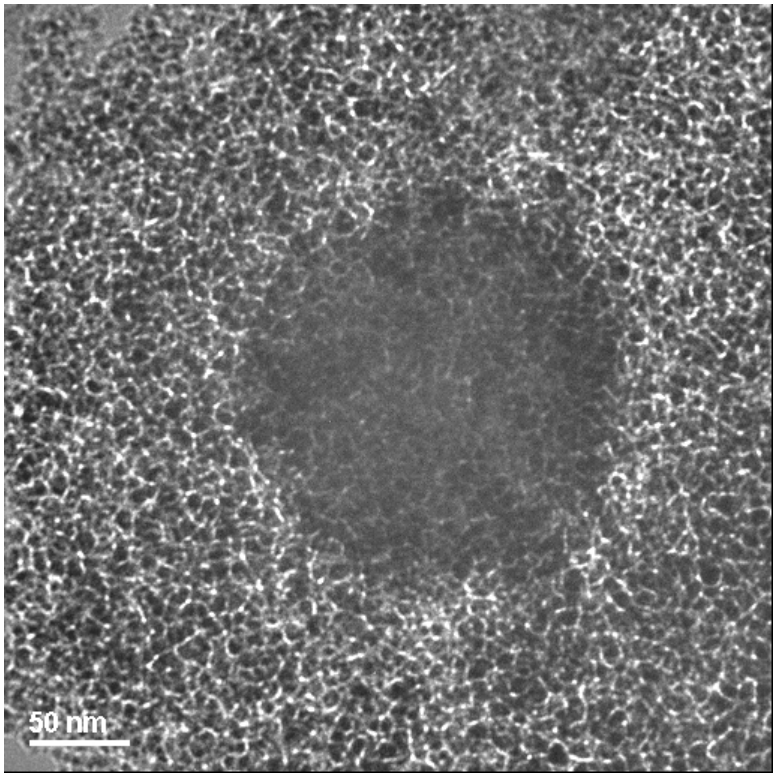

Fig. S2 TEM images of sample 10 h, showing a spherical morphology and typical size of the zeolite crystallites that are embedded in a mesoporous matrix.

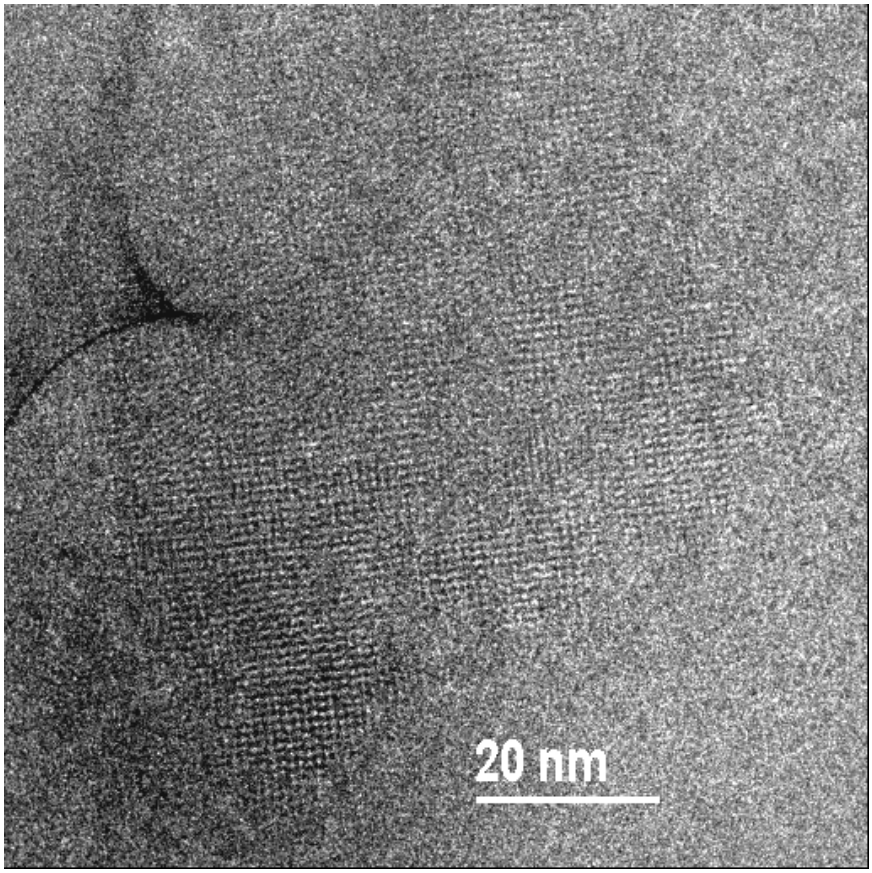

Fig. S3 HRTEM image (sample 3 h) of a zeolite crystallite showing 2D lattice fringes. The principal d-spacings of the lattices are 1.04 and 0.99 nm, corresponding to (200) and (020) interplane distances in the ZSM-5 unit cell (a = 2.01 nm; b = 1.97 nm; c = 1.31 nm). The foam-like image contrast pattern surrounding the crystallite disappears at the current focus condition.

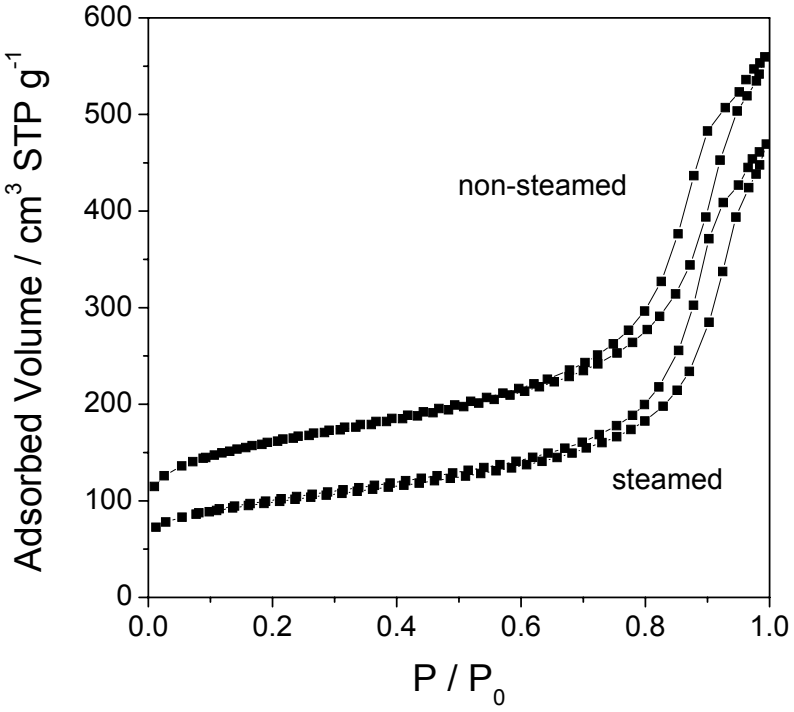

Fig. S4 N_2 adsorption/desorption isotherms of sample 15 h before and after hydrothermal treatment at 800 °C with 20 % water in helium for 2 h. There is no significant difference between the mesopore structures of the two samples. Micropore volume drops slightly after the treatment.

Comparison between traditional MFI zeolite synthesis and our method:

The traditional synthesis of MFI-type zeolite is based on the sol-gel method. Jacobsen *et al.* described a typical procedure involving TPAOH as a structure-directing agent (***JACS***, 2000, 122, 7116-7117). A clear synthesis solution containing Si and Al precursors, NaOH, TPAOH and water was heated to 180 °C in an autoclave for 3 days to obtain well developed ZSM-5 crystals. As reported elsewhere, the synthesis temperature can be as low as 100 °C and crystallization for 1 day.

Our synthesis is a solid-phase crystallization process, where liquid water phase and the solid are not in direct contact. (Fig. S5) The solid phase is placed in an open container standing inside the autoclave, and a small amount of water was placed at the bottom before heating up. At the reaction temperature (130 °C) water vapour is formed and solid samples are in contact with steam. After 3 to 15 hours of crystallization MFI zeolite crystals are formed with different sizes in the solid sample. Furthermore, the composition is different from the traditional sol-gel synthesis since no NaOH is used and the water content is lower.

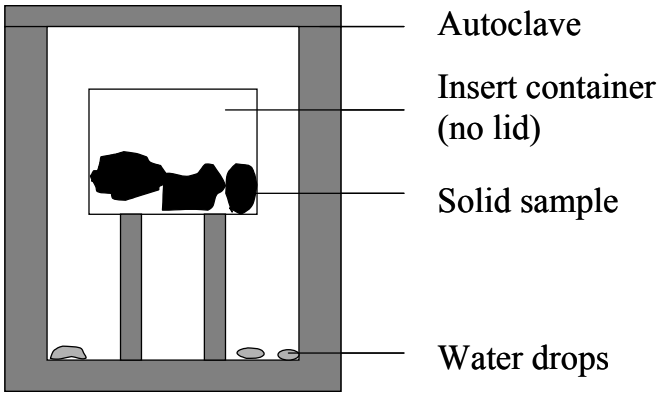

Fig. S5. A specially designed autoclave for TUD-C synthesis.