Supporting information

Mg²⁺-derived mesoporous ultra-high silica twelve-membered-ring basic zeolites: straightforward synthesis and catalytic performance

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Figure S1 XRD patterns of as-calcined samples, A) high silica zeolite with different SiO_2/Al_2O_3 , a) 300, b) 350, c) 500, (d) 550, synthesis condition: the crystallization time: 14 d, the pH of crystallization is 12.5; B) as-calcined samples with SiO_2/Al_2O_3 of 300 on different crystallization time, a) 6d, b) 18d; C) a) sample prepared by base co-hydrolysis, samples synthesized at different initial pH value of hydrolytic process, b) pH > 1.0, c) pH <0.7; D) Mg-free zeolite synthesis in different conditions, a) $SiO_2/Al_2O_3=200$, pH = 12.5 (pH of crystallization), b) $SiO_2/Al_2O_3=800$, pH = 13.02 (pH of crystallization).



Figure S2 N_2 adsorption-desorption isotherms and the corresponding pore-size distribution of Mg²⁺-derived mesoporous ultra-high silica zeolites.



Figure S3 SEM images for as-calcined samples: a) Mg-BEA(200), b) Mg-BEA(800), c) Mg-MTW(700) and d) Mg-MTW(1000).



Figure S4 Catalytic stability of Mg²⁺-derived mesoporous ultra-high silica zeolites in Knoevenagel condensation of benzaldehyde with ethyl cyanoacetate.



Figure S5 Framework topologies of ZSM-12 (MTW) and beta (polymorph A, BEA, and polymorph B, BEB) viewed along the straight 12R channels.

Figure S5 depicts the framework topologies of ZSM-12 (MTW) and Beta (polymorph A, BEA, and polymorph B, BEB). Although BEB is not officially used for the Framework Type Code (FTC) in the database of the Structure Commission of the International Zeolite Association (IZA-SC) (Ch. Baerlocher, L.B. McCusker and D.H. Olson, Atlas of Zeolite Framework Types, Sixth Revised Edition, Elsevier, Amsterdam, 2007. Available at: http://www.iza-structure.org/databases), here it is just used for convenience in order to present the similarity in the framework structure of ZSM-12 and Beta. ZSM-12 and beta possess very similar topology in which their a–c projection viewed along and perpendicular to the 12R straight channels is the same as reported by Lobo et al. (*R.F. Lobo, M. Tsapatsis, C.C. Freyhardt, I. Chan, C. Y. Chen, S.I. Zones and M.E. Davis, J. Am. Chem. Soc., 1997, 119, 3732–3744) and Kubota et al.* (*Y. Kubota, T. Honda, J. Plevert, T. Yamashita, T. Okubo and Y. Sugi, Catal. Today, 2002, 74, 271–279*). This indicates that beta seeds would be able to provide a specific growth surface for the crystallization of the MTW phase through their structural similarity, which therefore makes it understandable that MTW appearing as the first crystalline phase at t < 6 d in this work has transformed into BEA upon the further hydrothermal treatment of the reaction mixture.