Continuous Flow Synthesis of ZSM-5 Zeolite on the Order of Seconds

Toru Wakihara[1], Zhendong Liu,[1] Kotatsu Okabe,[1] Chokkalingam Anand,[1] Yasuo Yonezawa,[1] Jie Zhu,[1] Hiroki Yamada,[1] Akira Endo,[2] Yutaka Yanaba,[3] Takeshi Yoshikawa,[3] Koji Ohara,[4] Tatsuya Okubo[1]

[1]*Department of Chemical System Engineering, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo, Japan.*
[2]*National Institute of Advanced Industrial Science and Technology (AIST), Ibaraki 305-8565, Japan.*
[3]*Institute of Industrial Science, The University of Tokyo, 4-6-1 Komaba, Meguro-ku, Tokyo, 153-8505, Japan.*
[4]*JASRI/Spring-8, Kouto 1-1-1, Sayo-gun, Hyogo 679-5198, Japan.*

E-mail: *wakihara@chemsys.t.u-tokyo.ac.jp*

Zeolites have typically been synthesized *via* hydrothermal treatment, a process designed to artificially mimic the geological formation conditions of natural zeolites. This synthesis route, typically carried out in batch reactors like autoclaves, takes a time so long (typically, on the order of days) that the crystallization of zeolites had long been believed to be very slow in nature. Long periods of hydrothermal treatment also cause a burden on both energy efficiency and operational costs. Recently, we have reported the ultrafast syntheses of a class of industrially important zeolites within several minutes.[1,2] Further shortening the crystallization time to the order of seconds would be a great challenge but can significantly benefit the mass product of zeolites as well as the fundamental understanding of the crystallization mechanism.

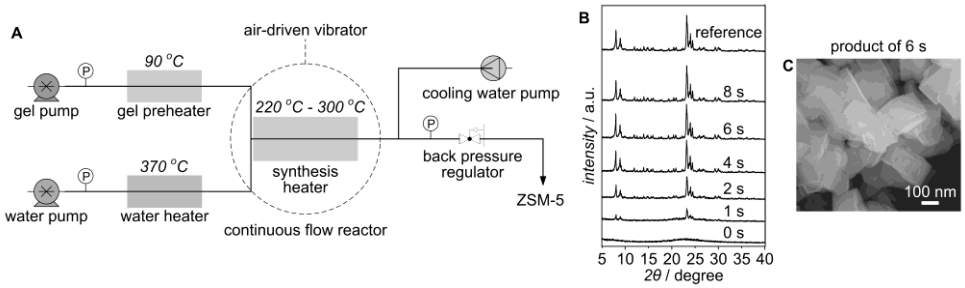

Fig. 7. (A) Flowchart for the continuous flow synthesis. (B) XRD patterns for the the products synthesized over different periods at 260 °C. (C) SEM image for the ZSM-5 product synthesized in.

We present herein a continuous flow method for the synthesis of ZSM-5 using pressurized hot water with extremely high temperature (370 °C) as the heating medium. Direct mixing of synthesis precursor and the pressurized hot water in a millimeter-sized continuous flow reactor could result in immediate heating up to high temperatures (Fig. 7A); consequently, the crystallization of ZSM-5, from amorphous state to full crystallinity, proceeded to completion at a remarkably fast rate in a system without the addition of any seed. The well-tuned synthesis precursor, obtained by aging the initial aluminosilicate gel at 90 °C for a certain period, triggered the nucleation and ensured the formation of ZSM-5 without any byproduct at extremely high temperatures. Consequently, the crystallization rate surpassed the decomposition rate of OSDA because of the creation of a favorable environment for ultrafast crystallization. When a gel aged for 16 h was employed, fully crystalline ZSM-5 was obtained after a synthesis for 6 s (Fig. 7B). SEM image in Fig. 7C shows that the ZSM-5 synthesized after 6 s exhibited well crystallized facets.

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[2] Liu, Z.; Wakihara, T.; Nomura, N.; Matsuo, T.; Anand, C.; Elangovan, S. P.; Yanaba, Y.; Yoshikawa, T.; Okubo, T. *Chem. Mater.* **2016**, 28, 4840