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Note

Fabrication of Zeolite A Rods with Irregular Macropores by Self-assembly of Zeolite A Microcrystals Using Microwave-assisted Hydrothermal Synthesis

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Zeolite A rods by self-assembly of zeolite A microcrystal were successfully synthesized by microwave-assisted hydrothermal synthesis. The average size of zeolite crystals consisting of self-assembling materials was about 300 nm and the length of zeolite rods was in the range of 15—30 µm.

Keywords zeolite rods, A-type zeolite, microwave synthesis

Fabrication of zeolite structures in other geometries such as thin zeolite films,[1,2] micro-macroporous zeolite structures[3,4] and long zeolite fibers[5] has found widespread interest due to its promise to improve the separation and catalysis efficiency of zeolite materials, and otherwise has been used to new applications such as that in microelectronic devices.

Up to now, a series of zeolite materials with mesoporous structure by using self-assembly of zeolite nanocrystals in conventional synthesis have been developed, such as MAS-5,[6] MAS-7[7] and MTS-9.[4] Microwave hydrothermal synthesis, as a fresh highly efficient heating way, has been increasingly focused, particularly in zeolite synthesis.[2] In this work, we report the fabrication of zeolite rods with irregular macropores by self-assembly of zeolite A microcrystals using microwave-assisted hydrothermal synthesis.

A nutrient solution with a molar composition ratio of $3Na_2O$: $2SiO_2$: $1Al_2O_3$: $150H_2O$: 60EtOH : 0.01PVA was prepared by first dissolving $Al(OH)_3$ (A.R., 99.9%) in a 6 mol/L NaOH solution and then introducing a SiO_2 sol (25 wt% in water) under intense stirring at room temperature. After a further half hour of stirring, an amount of ethanol (A.R., 99.9%) and polyvinyl alcohol (A.R., M_r=6000) was respectively added dropwise. The resulting solution (30 mL) was transferred into a Teflon vessel in which the α-Al_2O_3 substrate (the diameter in 24 mm, the thickness in 1.5 mm, and the porosity in 60% as well as average pore size in 0.3 µm), was vertically mounted in advance. The Teflon vessel was then attached to a condenser and heated at a power of 100 W for 20 min in a microwave oven under reflux condition. The support after synthesis was washed several times by deionized water until neutrality and dried at room temperature.

The formation of self-assembling materials was confirmed by X-ray diffraction analysis (XRD) using a Rigaku Rotaflex D/MAX-C powder diffractometer with Cu Kα (λ=0.154 nm) radiation, with operation condition at 40 kV and 30 mA. Electron micrographs were recorded with an XL-30 ESEM TMP (PHILIPS) scanning electron microscope.

Figure 1 shows the XRD patterns of the macroporous self-assembling materials. It can confirm that the

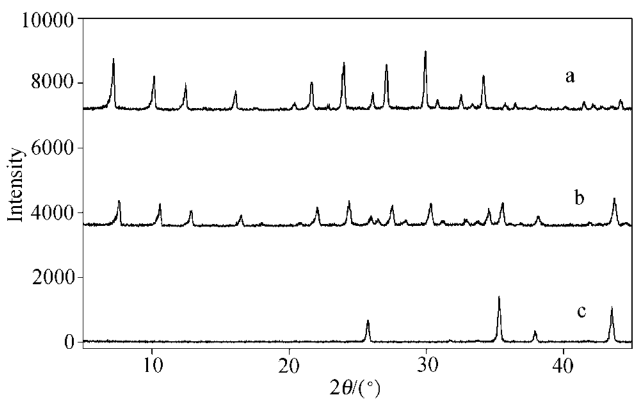

Figure 1 XRD patterns of the zeolite rods by self-assembly of zeolite A microcrystals. a: zeolite A powder; b: zeolite rods; c: the substrate.

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self-assembling materials consist of zeolite A microcrystals. In comparison of the XRD pattern, there is not obvious discrepance between the zeolite A powder and the self-assembling materials, suggesting that the latter is composed of free-oriented zeolite crystals.

The SEM images of the self-assembling materials are shown in Figure 2. The yield of the self-assembling materials is remarkably high, demonstrating that the approach of self-assembly forming zeolite rods is practicable. The self-assembling materials possess many irregular macropores which are fabricated by self-assembly of zeolite microcrystals. The average size of zeolite crystals consisting of self-assembling materials is about 300 nm (see Figure 2-c, d). The length of zeolite rods is in the range of 15—30 μm (see Figure 2-a). Interestingly, the studied results also show that there are not zeolite rods but zeolite membrane grown on substrate in the absence of polyvinyl alcohol, suggesting that the presence of polyvinyl alcohol likely plays an important role for making the self-materials.

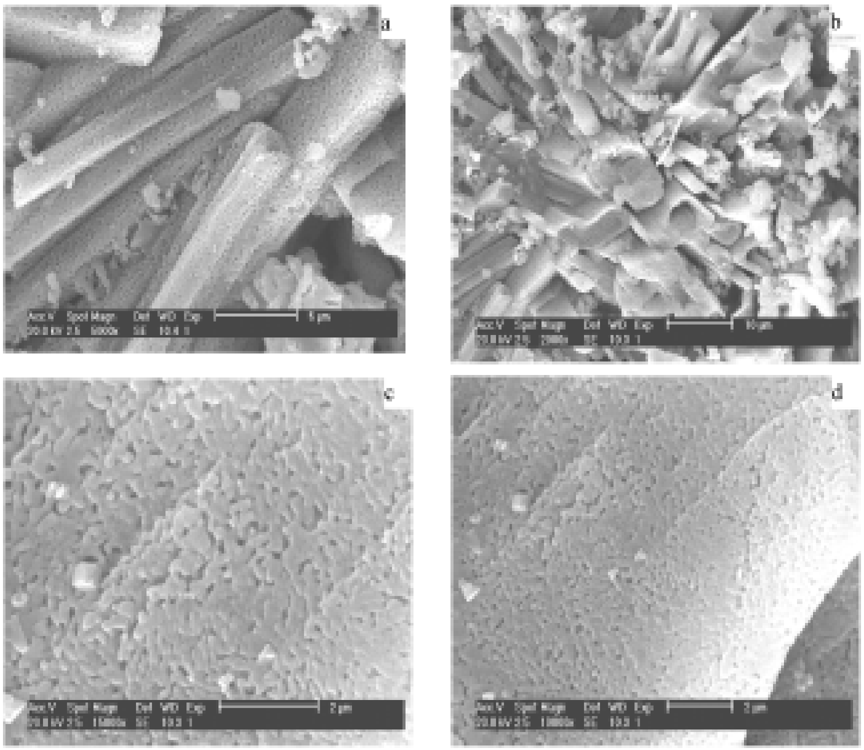

Figure 2 SEM images of zeolite rods. a. top view; b: cross-section view; c, d: high resolution (top view).

References

1 Xu, X. C.; Yang, W. S.; Liu, J.; Chen, X. B.; Lin, L. W.; Stroh, N.; Brunner, H. *Chem. Commum.* **2000**, 603.

2 Xu, X. C.; Yang, W. S.; Liu, J.; Lin, L. W. *Adv. Mater.* **2000**, *12*, 195.

3 Huang, L. M.; Wang, Z. B.; Sun, J. Y.; Miao, L.; Li, Q. Z.; Yan, Y. S. Zhao, D. Y. *J. Am. Chem. Soc*. **2000**, *122*, 3530.

4 Xiao, F. S.; Han, Y.; Yu, Y.; Meng, X. J.; Miao, Y.; Wu, S. *J. Am. Chem. Soc*. **2002**, *124*, 888.

5 Jung, K. T.; Hyun, H. H.; Shul, Y. G.; Kim, D. S. *Zeolites* **1997**, *19*, 161.

6 (a) Zhang, Z. T.; Han, Y.; Zhu, L.; Wang, R. W.; Yu, Y.; Qiu, S. L.; Zhao, D. Y.; Xiao, F. S. *Angew. Chem., Int. Ed.* **2001**, *40*, 1258.
(b) Zhang, Z. T.; Han, Y.; Zhu, L.; Wang, R. W.; Yu, Y.; Qiu, S. L.; Zhao, D. Y.; Xiao, F. S. *J. Am. Chem. Soc*. **2001**, *123*, 5014.

7 Han, Y.; Xiao, F. S.; Wu, S.; Sun, Y.; Meng, X.; Li, D.; Lin, S.; Deng, F. *J. Phys. Chem. B* **2001**, *105*, 7963.

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