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A two-phase segmented microfluidic technique for one-step continuous versatile preparation of zeolites

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HIGHLIGHTS

- One-step versatile continuous synthesis of zeolites was implemented.
- A two liquid phase segmented microfluidic device was used for the synthesis.
- The silica and alumina solutions were mixed in the device to form the hydrogels.
- Zeolite A with adjustable sizes could be synthesized by changing the flow rates.

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ABSTRACT

A technique for one-step versatile continuous synthesis of zeolites in a two liquid phase segmented microfluidic device was developed. This was conducted by forming the zeolite synthesis hydrogel micro-droplets in a continuous paraffin phase through pumping a silica solution and an alumina solution respectively into two closely-packed stainless steel capillaries positioned in the axis of a PTFE outer tube, followed by crystallization in the PTFE tube at 90 °C. The synthesis of zeolite A was carried out as an example to demonstrate the feasibility of this technique. The influences of the dimensions of the tubes, the reaction time and the flow rate ratios of silica and alumina solutions were examined. The synthesized zeolite A crystals were characterized by X-ray diffraction, Fourier transform infrared spectroscopy, and scanning electron microscopy. The particle size of products can be controlled in the range of 0.9–1.5 μm by changing the flow rate ratios of silica and alumina solutions. The sizes of the hydrogel segments determine the mixing efficiency and control the crystallization process. Decrease in the dimension of outer tube or ultrasonic radiation can decrease the sizes of hydrogel segments, leading to improvement of the mixing efficiency, intensification of the crystallization process, and subsequent formation of high crystalline zeolite A crystals. Furthermore, metastable transformation of zeolite NaA to sodalite occurs with prolongation of the reaction time from 30 to 120 min. The technique developed in this paper can be applied in the continuous synthesis of other kinds of zeolites.

1. Introduction

Zeolites, crystalline aluminosilicates with unique pore sizes and high capacity of ion exchange, have a wide range of applications in detergent, drying, adsorption, gas separation, catalysis, fuel cells and so on [1–5]. They are conventionally synthesized in batch reactors, though there are several limitations associated with the batch processing, including inhomogeneous reactant and temperature distributions, inefficient mixing, and product variations from batch to batch. Up to now, few reports about continuous and semi-continuous synthesis of zeolite crystals in stirred tanks or tubular reactors have been given, which are usually carried out by first mixing the silica and alumina solutions to prepare the synthesis hydrogel, then transferring the mixture into heated stirred tanks or tubular reactors for the crystallization [6–9]. It would be more favorable if the silica and alumina solutions can be mixed directly in the tubular reactors, thus eliminating the huge tank for preparing the zeolite synthesis solutions. However, mixing the two solutions directly in the tubular reactor may result in clogging because of rapid formation of viscous hydrogels.

Microfluidic reactors, with excellent heat transfer and rapid mixing properties, have been widely applied in preparation of inorganic and polymer particles and organic synthesis [10–15]. Especially, two-phase liquid segmented microfluidic patterns for hydrothermal synthesis of particles could keep the precursors and solid products away from the tube inner walls to avoid clogged channels [16–18], providing a feasible and simple strategy for

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