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Article

Alcohol-assisted synthesis of high-silica zeolites in the absence of organic structure-directing agents

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ABSTRACT

In this work, we show for the first time that high-silica zeolites (MFI, TON, MTT, and *MRE) could be synthesized from a combined strategy of both zeolite seeding and alcohol filling in the absence of organic structure-directing agents (OSDAs). High-silica ZSM-5 zeolites with Si/Al ratios ranging from 38 to 240 (TF-Al-ZSM-5) could be synthesized via this route. The key to the success of this technique was the employment of an aluminosilicate precursor with a fully 4-coordinated aluminum species as the initial source, wherein the rearrangement and condensation of the silicate species, rather than the aluminate species, occurred during zeolite crystallization. In addition, heteroatoms, such as Fe and B, could be incorporated into the zeolite frameworks. Catalytic tests for the methanol-to-propylene (MTP) reaction exhibited good catalytic performance for TF-Al-ZSM-5, which was comparable to that of the aluminosilicate ZSM-5 zeolite synthesized with OSDAs. Hence, this method offers viable opportunities for the industrial production and catalytic application of high-silica zeolites in the future.

1. Introduction

Zeolites, particularly high-silica zeolites, have been extensively applied in the field of catalysis for decades owing to their excellent thermal and hydrothermal stabilities, large micropore volumes, high surface areas, and uniform microporous channels [1–27]. For example, high-silica aluminosilicate ZSM-5 zeolite is widely used in the methanol-to-propylene (MTP) reaction [22–24], and high-silica aluminosilicate ZSM-48 zeolite is an efficient catalyst for the hydroisomerization of n-alkanes [24–27]. Generally, high-silica zeolites are synthesized in the presence of organic structure-directing agents (OSDAs), such as quaternary ammonium cations and amines, which direct the assembly pathway and ultimately fill the zeolite micropores [8–27]. The employment of these OSDAs not only increases the cost but also produces a large amount of polluted water. In addition, the zeolite framework sometimes would deteriorate during the removal of organic templates due to high temperature calcination.

In the past years, great efforts have been made to develop

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