Molecular Sieve Synthesis using Imidazolium Structure Directing Agents

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Abstract

The synthesis of high-silica molecular sieve SSZ-70 is investigated through a guest/host study of imidazolium structure directing agents (SDAs). The original borosilicate synthesis is extended to pure-silica and aluminosilicate compositions using six imidazolium SDAs. Physical characterization using powder X-ray diffraction (XRD), ^{29}Si solid-state NMR, electron microscopy, thermogravimetric analysis, and nitrogen adsorption shows SSZ-70 to be layered with similarity to MCM-22 (MWW). Aluminum-containing SSZ-70 is evaluated for catalytic activity using the constraint index (CI) test and shows a similar cracking rate to SSZ-25 (MWW structure). Distinct differences in CI as a function of time on stream are observed between MWW and SSZ-70 materials. Additional molecular sieve phases observed in this guest/host study included Theta-1 (TON), ZSM-5 (MFI), ZSM-23 (MTT), ZSM-12 (MTW), Beta, Mordenite (MOR), CIT-5 (CFI), SSZ-16 (AFX), and SSZ-35 (STF).

Attempts to synthesize Beta enriched in chiral polymorph A are investigated in a second guest/host study using five chiral imidazolium SDAs. Two SDAs successfully gave Beta, but no enrichment in polymorph A is observed. The remaining SDAs do not direct the formation of any molecular sieve phases. Molecular modeling indicates both SDAs occupy the straight [100]/[010] 12 membered ring (MR) pores of Beta. In this configuration, no chirality could be projected across the [001] fault planes and this offers an explanation for not observing enrichment. Modeling shows careful consideration must be given to efficiently filling the entire void volume when large SDAs are used. Additional molecular sieve phases observed in the guest/host study are EU-1 (EUO) and MOR.

Finally, attempts to synthesize novel materials using supramolecular SDAs are described. Supramolecular SDAs are created through adamantyl/β-cyclodextrin inclusion complex formation. Both 2:1 and 1:1 inclusion complex stoichiometries are attempted. Significant cyclodextrin degradation occurs at temperatures above 90°C and no structure-directing effect can be attributed to the cyclodextrin. Molecular sieve phases observed in the study are SSZ-16 (AFX), MOR, B-SSZ-13 (CHA), VPI-8 (VET), and SSZ-24 (AFI).

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