

Ionic-Liquid-Templated Synthesis of 10-Membered Ring Zeolites with MFI and TON Morphologies

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Tailoring zeolite structures has been demonstrated to be a significant means to improve the catalytic performance in industrial applications. We elucidated the effect of tetraethyl orthosilicate (TEOS) hydrolysis before the hydrothermal treatment on the final zeolite phase selectivity in the ionic liquid-templated synthesis of 10-membered ring zeolites (MFI or TON type zeolite) (Scheme 1). X-ray diffraction (XRD) patterns of samples synthesized using multiple silica sources indicated the zeolite-structure-determining role of the TEOS hydrolysis process. Systematic investigation of the synthesis solution revealed that the dissolved silica concentration in the synthesis solution, which is effectively controlled by varying the TEOS hydrolysis temperature and addition rate, is a crucial determinant of the final zeolite phase selectivity during the hydrothermal treatment towards TON or MFI. Both the electrospray ionization mass spectrometry (ESI-MS) and attenuated total reflectance-Fourier transform infrared spectra (ATR-FTIR) confirmed the comparable compositions of the supernatants before hydrothermal treatment regardless of the TEOS addition rate and hydrolysis temperature. It is inferred that heterogeneous nuclear growth occurred and produced different metastable structures for various zeolite types. ATR-FTIR and XRD analyses of the solids recovered after different crystallization durations revealed the chemical heterogeneity of the precursor nanoparticles. The results are supported by theoretical modelling of silica polymerization via reaction ensemble Monte Carlo (REMC) and building unit analyses using Composite Building Unit (CBU) prediction. The results are valuable for understanding the nucleation mechanism in zeolite crystallization.

Scheme 1. The proposition of [BMIM]Br-directed Synthesis of MFI and TON zeolites via Controlling TEOS Hydrolysis.

