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Kinetics and Mechanisms of Zeolite Crystallization at Hyperalkaline Conditions

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The predicted precipitation of zeolites in geologic barrier systems for radioactive wastes due to the alkaline alteration of bentonite may result in the modification or loss of favorable physicochemical properties of the bentonite as a suitable barrier material. Zeolites formation is typically preceded by an amorphous precursor, the transformation of which is seen as the rate-controlling step. However, the structure of the precursor phase and the rates and mechanisms by which it transforms into crystalline zeolites are poorly understood. In this study, we investigated the rates and mechanisms of zeolite crystallization from solutions.

Batch synthesis experiments were carried out over a range of solution compositions (Si/Al = 0.1 to 8.0), pH (9.5 to 13.5) and temperature (25C to 90C) conditions in order to clarify the effects of these parameters on zeolite crystallization. Solid products were characterized using XRD, SEM-EDX, FTIR spectroscopy, Raman spectroscopy and MAS NMR spectroscopy.

Zeolite crystallization proceeds by the rapid formation of an amorphous precursor phase, followed by the slower transformation of this precursor into crystalline zeolite. Depending on the Si/Al ratio of the parent solution, the species of zeolite may vary. At Si/Al >1, Faujasite forms slowly, whereas for Si/Al <1, Zeolite A forms more rapidly. Higher pH and temperatures favor transformation.

Morphological information from SEM shows intimate physical relationship between crystalline zeolites and the amorphous precursor phase. Spectroscopic results from FTIR, Raman and MAS NMR indicate that ring structures are present in both amorphous and crystalline phases, indicating structural similarity between the two phases. These data may suggest that amorphous phases transform directly into crystalline zeolites. The activation energy of crystallization suggests that solid-state processes occur alongside dissolution of the amorphous phase in order for the transformation of the amorphous phase into crystalline zeolite to proceed.

Keywords: zeolite, mechanisms, transformation, spectroscopy