Impact of cations on (un)desirable zeolite transformations at high pH

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High alumina zeolites are typically synthesized hydrothermally in hyperalkaline conditions. Postsynthetic alkaline treatment induces partial or complete dissolution, followed by nucleation and growth of new (zeolite) products. Major factors controlling such zeolite transformations are temperature, alkalinity and the presence of specific cations in the zeolite and the alkaline medium. While material scientists exploit zeolite transformations to design novel zeolite materials, zeolite stability in hyperalkaline media is of high interest to geoscientists, evaluating long-term stability of natural zeolites used as reactive barrier in concrete based disposal strategies for nuclear waste.

This contribution discusses the role of alkali metal cations on FAU (faujasite) type zeolite transformations in 1 M hydroxide solutions under autogeneous pressure at 95°C or lower. Liquid and solid phase analysis was performed as function of contact time, with ICP-AES, NMR, SEM and XRD. Although exposure of FAU to KOH nowadays is a standard recipe to synthesize chabazite (CHA), few studies dealing with the transformation mechanism and kinetics are available. Our systematic study revealed, among others, that the presence of K⁺ ions is crucial for the conversion of FAU to CHA. Identical transformation conditions yielded ABW, FAU, MER and ANA frameworks by substituting KOH with LiOH, NaOH, RbOH, and CsOH respectively. [1] In addition to cation identity, other important variables determining the outcome of the transformations were the Si/Al ratio of the initial FAU framework, and the solid/liquid ratio. Furthermore, CHA was obtained by contacting FAU with $K^{\scriptscriptstyle +}$ rich young cement water (YPW, pH 13.5) at 60°C, illustrating its potential use in such hyperalkaline conditions. In these conditions, a minor fraction of zeolite with MER (merlinoite) topology was detected in addition to CHA. The MER framework has buckled 8-rings of MER that form a perfect nest for K⁺.[2] Currently, the relationship between CHA and MER formation, starting from FAU, is investigated and compared to direct syntheses from amorphous sources. The hypothesis is explored that the FAU framework structure directs the CHA formation, as CHA was observed to nucleate on the FAU (111) crystal faces.

[1] Van Tendeloo *et al* (2013) *Chem. Commun.* **49**, 11737-11739 [2] Skofteland *et al* (2001) *Microporous Mesoporous Mater.* **43**, 61-71