## An NMR view of nucleation theory for zeolites crystallization

M. HAOUAS, L. VAN TENDELOO, C. E. A. KIRSCHHOCK, J. MARTENS, E. BREYNAERT AND F. TAULELLE\*

Center for Surface Chemistry and Catalysis, Kasteelpark Arenberg 23, 3001 Heverlee, Belgium Tectospin, Institut Lavoisier, University of Versailles St Q.Y., 45 Avenue des Etats-Unis, 78035 Versailles Cedex France \*correspondence: francis.taulelle@biw.kuleuven.be

Zeolites nucleation understanding has been blurred by the successful usage of gel synthesis. Though since about 40 years zeolites have been known to be synthesized from homogenous liquids. The industrial successes of gel synthesis of ZSM-5 and silicalite has inhibited research of zeolites from full homogenous liquids.

Recently, hydrated silicates ionic liquids (HSIL) have been used to synthesize zeolites.<sup>[1]</sup> They are hyperbasic media, with a number of water limited to coordination of ions.

With such silicated systems all stages of nucleation occur in very slow motion. Therefore, crystallization of zeolites can be investigated with all the tools related to homogeneous liquids. NMR has been successfully used to monitor all the steps of silicalite from sols.

In this presentation, a general model of nucleation will be exposed, unifying zeolite crystallization with all other nucleation schemes, from organic molecules to proteins and other inorganic crystals. Zeolites from HSIL and from sols allow to unfold, in slow motion all stages of nucleation. At crystallization formation of an anisotropic lattice of connecting interactions, generally called bonds, is differentiated. Such differentiation between weak bonds and strong bonds allows to unify molecular and covalent crystallization schemes. The timing of each stage leads to move continuously from classical nucleation theory to two-step nucleation schemes.

[1] Van Tendeloo L, Haouas M, Martens JA, Kirschhock CEA, Breynaert E, Taulelle F: Zeolite synthesis in hydrated silicate ionic liquids. *Faraday Discussions* 2015, DOI 10.1039/C4FD00234B. [2] Petry DP, Haouas M, Wong SCC, Aerts A, Kirschhock CEA, Martens JA, Gaskell SJ, Anderson MW, Taulelle F: Connectivity Analysis of the Clear Sol Precursor of Silicalite: Are Nanoparticles Aggregated Oligomers or Silica Particles? *J Phys Chem C* 2009, 113:20827–20836.