INSIGHTS INTO THE FORMATION OF ZEOLITES BY STUDYING OF THE REACTION PRECURSORS

Ana Palčić
Laboratory for the Synthesis of New Materials, Department of Materials Chemistry,
Ruđer Bošković Institute, Bijenička 54, 10000, Zagreb, Croatia
E-mail: ana.palcic@irb.hr

ABSTRACT

Zeolite materials design is governed/prompted by the demands ensuing from the technological challenges as well as from the growing needs for enhancing the environmental protection and increasing the energy efficiency and sustainability. New demands require fine control of zeolite physicochemical properties and the preparation of customized materials. Understanding the mechanism of zeolite synthesis is the precondition for efficient and rational design of desired final product. In the past sixty years vast number of studies on the matter has been performed. However, there are many points which are still not fully elucidated. Therefore deeper insights of atomic scale reactions leading to zeolite formation are indispensable. Herein, the potential to control zeolite crystal size and morphology will be addressed in terms of time and spatial consequence of the nucleation events.

The objective of present study is to correlate the nucleation process and crystallization pathway with the physicochemical properties of the zeolite. The study was performed on low silica zeolite systems – framework types LTA and EMT. Special attention was devoted to the study of the precursors. The primary results based on XRD, SEM, TG, positron annihilation lifetime spectroscopy and laser light scattering analyses show that the zeolite A crystals begin to grow at various stages of the reaction indicating that the nuclei are inhomogeneously distributed within gel particles. The nuclei distribution within the gel particles can be affected by the initial gel aging. In addition, the number of structured units assigned to viable nuclei gets higher as the gel aging time is increased. Further insights were obtained by analysis of EMT gel precursor particles by high resolution TEM and electron tomography. The latter study revealed the core-shell structure of the initial gel particles. The inception of zeolite nucleation is localized in the shell part of the gel particle. Consequently, the external surface area of the primary gel particles plays an important role in the nucleation process and directs the overall kinetics of zeolite formation.

The set of experimental results clearly indicate that cautious engineering of the initial gel particles can lead to the preparation of zeolite crystals with targeted size, uniform crystal size distribution and preferred morphology. Moreover, the proposed mechanism could be generalized for aluminosilicate zeolites formed from alkali-metal-rich hydrogel systems.

Keywords: zeolite nucleation, crystallization, crystal size, morphology

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