Peering into the Black Box of Zeolite Crystallization:
New Methods to Identify Mechanisms of Growth
and Tailor Physicochemical Properties for Catalytic Applications

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Crystal engineering is a broad area of research that encompasses the design and optimization of materials for diverse applications in fields spanning from energy to medicine. The ability to selectively control crystallization to achieve desired material properties requires detailed understandings of the thermodynamic and kinetic factors regulating crystal nucleation and growth. Combining this fundamental knowledge with innovative approaches to tailor crystal size, structure, and morphology can lead to materials with superior properties beyond what is achievable by conventional routes.

In this talk I will discuss how zeolites form by two mechanisms of crystal growth: (1) classical pathways involving 2-dimensional layer nucleation and advancement on crystal surfaces through monomer addition; and (2) nonclassical pathways, termed *crystallization by particle attachment* (CPA), involving the formation of metastable precursors that play a direct role in crystal nucleation and growth.

Our group uses atomic force microscopy (AFM) to investigate crystallization *in situ* under solvothermal conditions. This allows us to capture time-resolved dynamics of zeolite surface growth, thereby probing complex pathways of crystallization. To this end, we have identified unique modes of growth that expand our understanding of nonclassical crystallization. We have coupled these studies with parametric investigations of zeolite synthesis, focusing on commercially-viable methods to prepare zeolites in the absence of organic structure-directing agents, which poses numerous challenges (e.g., polymorphism).

An additional design approach that will be discussed is the use *modifiers* to control zeolite size and morphology. Modifiers are molecules or macromolecules that interact with specific surfaces of crystals and regulate anisotropic growth rates. We use this technique to generate zeolite catalysts with well-defined properties for studies aimed to elucidate structure-performance relationships in reactions such as methane upgrading and methanol to hydrocarbons. Our results collectively reveal that the rational design of zeolites can dramatically improve catalyst performance.

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