# SYNTHESIS AND CATALYTIC PROPERTIES OF HIERARCHICALLY STRUCTURED ZEOLITE CATALYSTS WITH INTRACRYSTALLINE MACROPORES

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Zeolites belong to the most important heterogeneous catalysts. They are widely applied in crude oil refining, petrochemistry, fine chemistry, as well as in environmental applications. A unique feature of zeolites is their well-ordered micropore system with pore diameters similar to the dimensions of molecules. These small pores give rise to the shape selective properties of zeolite catalysts. However, the diffusion of molecules to and from the active sites confined within the micropores is very slow, which often leads to diffusion limitations. These diffusion limitations result in reduced utilization of the zeolite crystal and can also lead to reduced selectivity or lifetime of zeolite catalysts. A nature-inspired approach to overcome such diffusion restrictions is the utilization of catalysts with an optimally designed, hierarchical structure. In nature, mass transport systems, such as trees or lungs, possess an optimized hierarchical architecture to reduce transport limitations across a wide range of length scales.[1] Adapting this approach to zeolites can be realized by including at least one additional system of larger pores interconnected to the zeolitic micropores. Hereby, hierarchical zeolites coul already demonstrate enhanced diffusion properties and, consequently, better catalytic performance.[3]

In order to prepare a truly nature inspired catalyst, a guided material design is crucial. Therefore, the transport pore system must exhibit an optimal porosity and the zeolitic domains in between the transport pores need to be small enough to eliminate local diffusion limitations. The pore size can be neglected, if it is larger than a certain minimum pore size, usually in the range of macropores or very large mesopores.[3] However, preparation approaches for hierarchical zeolites are often unguided and result mostly in materials containing relative small mesopores. In this contribution we introduce a synthesis approach for zeolite single crystals with intracrystalline macropores by a so-called inverse crystallization, which allows control over the porosity, pore size and wall thickness of the hierarchical zeolite (see Figure 1 b). This synthesis approach utilizes mesoporous spherical silica particles as a sacrificial template for the macropore formation during zeolite synthesis by steam-assisted crystallization. Furthermore, we show the effect of these additional intracrystalline macropores on the catalytic performance for the direct conversion of methanol to short chain olefins (MTO), with focus on coke formation and catalyst lifetime.

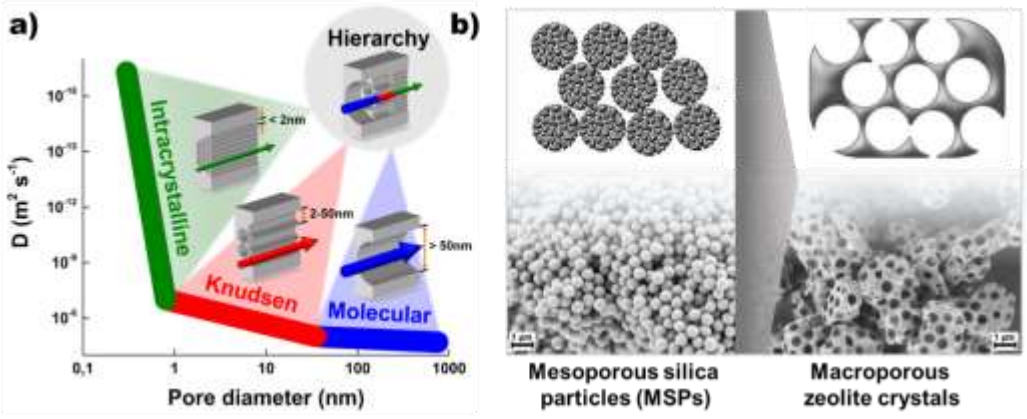


*Figure 1 – a) Diffusion coefficient over pore diameter and b) schematic representation of the inverse crystallization*

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