University of Maine
Department of Chemistry
Colloquium

Hierarchical nanofabrication of microporous catalysts with ordered mesoporosity

Prof. Wei Fan

Department of Chemical Engineering
University of Massachusetts, Amherst

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ABSTRACT: Due to the intrinsic ordered micropore structures and strong acid strength, zeolites are extensively used as heterogeneous acid catalysts in oil refining processes and chemicals synthesis. However, the micropore structures and high intrinsic activities frequently lead these materials to be subject to diffusion limitations that restrict reactant accessibility to the active sites on the interior surfaces of zeolites and inhibit the full utilization of zeolite catalysts. Nanofabrication of hierarchical zeolite catalysts with mesoporosity is a proven strategy for integrating shape selectivity provided by the intrinsic micropore structures and efficient mass transport facilitated by the mesopore structures. Considerable efforts have been devoted to the synthesis of zeolite catalysts with mesoporosity. Among the various approaches, the synthesis of hierarchical zeolites by the confined synthesis in hard templates holds exciting implications in term of creating ordered mesopore structures and controlling mesopore sizes. These advantages provide unique opportunities to design the hierarchical pore structures and quantitatively investigate the effects of mesopore structure on the catalytic performance of zeolite catalysts.

In this talk, I will demonstrate how a wide range of zeolite morphologies can be realized through such a confined growth within 3-dimensionally ordered mesoporous (3DOm) carbon with the pore size smaller than 40 nm. The carbon template was synthesized by replication of colloidal crystals composed of size-tunable silica nanoparticles. Confined crystal growth within the carbon template leads to size-tunable, uniformly shaped zeolite nanocrystals as well as 3DOm-imprinted single-crystal zeolite particles. In addition, novel crystal morphologies, consisting of faceted crystal outgrowths from primary crystalline particles have been discovered, providing new insights into constricted crystal growth mechanisms underlying the confined growth. The approach exhibits versatile abilities for controlling the mesoporosity of hierarchical zeolites from 5 nm to 10 nm. The mass transport in the hierarchical zeolites has been significantly improved, which can mostly be attributed to the shortened diffusion path length in the micropores. The catalytic performance of hierarchical zeolite catalysts for the reactions of bulky chemicals and biomass conversion will be discussed.

Host: Howard Patterson (howard.patterson@umit.maine.edu ; 581-1178)