559c Optimal Porosity Distribution in Nanoporous Catalysts

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We investigated the optimal distribution of porosity in a micro/mesoporous (nanoporous) catalyst. Given a heterogeneous catalyst with an imposed texture at the nano-scale, such as a zeolite or a mesoporous sieve (MCM-41, MCM-48, SBA-15, MSU, etc...) a question of interest is how to distribute large pores (macropores and large mesopores), so as to reduce diffusion limitations. These large pores may be inter-particle pores in a pellet or intra-particle channels of a broad pore size distribution. This problem is of great relevance to the practical use of rationally designed nanoporous catalysts, since the absence of a well-designed pore network structure at larger length scales might adversely affect the performance. By extension, this problem is also of relevance to chromatography, tissue engineering, and controlled release or take-up. Too few large pore channels in a big particle lead to severe diffusion limitations and an inadequate use of the carefully designed catalytic sites and nanoporosity, resulting in lower yields and lower selectivity to the desired product(s). Large pore channels facilitate access to the active sites, but reduce the amount of active catalyst. There clearly is an optimum porosity.

The way in which the porosity should be distributed is non-trivial. Our calculations demonstrate that hierarchical pore networks, somewhat reminiscent of networks in nature (e.g., the venal network of leaves), with a gradient in porosity from the outside to the inside of the catalyst particle are often better than a uniform porosity distribution. Calculations based on optimal control theory as well as computer simulations point to this. One compelling result is that the sub-class of self-similar networks of fractal dimension $2 < D < 3$ is better than a uniform network ($D = 3$) or no network of large channels at all ($D = 2$).

We shortly put this theoretical work in the context of the synthesis of hierarchically structured nanoporous materials, a field in which much progress is being made.