140a Multiscale Models for Diffusion-Controlled Selective Catalytic Oxidation in Nanopores

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Nanostructured membranes fabricated using anodic aluminum oxidation (AAO) and atomic layer deposition (ALD) have excellent characteristics for supporting heterogeneous catalysts and permit exquisite tailoring of the pore diameter and wall composition. This research is focused on the computational study of the selective catalytic oxidation of hydrocarbons inside nanoporous alumina membranes. Due to the short contact time, there is the potential to achieve high selectivity by avoiding over-oxidation. Our goal is to develop a model capable of predicting the transport and kinetic behavior inside the membranes and to propose an optimum pore design capable of maximizing the desired products.

Detailed atomistic molecular dynamics simulations were performed in pores with diameters ranging from 10 to 150 nm using ethane as a model molecule. For computational efficiency, the pore walls were represented using an analytical potential derived for smooth slit pores in combination with an explicit layer of the innermost oxygen atoms of the pore to provide a realistic model of pore roughness. Our results show that Knudsen diffusion is the dominant mass transfer mechanism for the system under the conditions of interest. Based on these results, a more coarse-grained model that simulates a boundary driven ensemble of particles in the Knudsen regime was developed to determine the number and location of hits between the particles and the wall, the residence time of the particles and the transport diffusivity. This model is the foundation for a hybrid model that includes simultaneous reaction and transport in the membranes. Also, a continuum reaction/diffusion model was used for the exploration of trends in product selectivity for the case of a homogeneous wall composition.