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Catalysis by Confinement, Electron Transfer and Lewis Acidity in Zeolites

The catalytic applications of zeolites are predominantly due to the Bronsted acidity of bridging hydroxide groups in the zeolite framework (Si-OH-Al), or due to extra-framework ions adjacent to the Al sites such as in copper-exchanged zeolites in the SCR of NO_x with ammonia. In contrast to these well-known catalytic processes, in this talk we will explore other aspects of zeolite catalysis that open new opportunities for applications at the industrial level.

The first example will explore catalysis by confinement through the specific case of NO oxidation over siliceous zeolites. We will see that the key to effective catalysis in this system depends on the ability of the 'pore space' to stabilize the transition state of this reaction by non-specific interactions and how this approach can be used to predict similar catalytic effects in microporous carbons and metal-organic frameworks.

The second example will examine electron transfer processes between adsorbents with low ionization potential (such as styrene) and the acid sites of high-silica zeolites. This information will be used to support the idea that in alkane activation over H-[Fe]zeolites, catalysis can proceed via electron transfer and through the formation of radical cation intermediates. Differences in activation energy and selectivity clearly indicate that the reactivity of these materials is inconsistent with acid catalyzed processes.

In the last example we will explore the use of Lewis-acid containing zeolites (Sn-, Zr- and Hf-beta) as catalysts for the Diels-Alder reaction. In particular, we will describe new processes for the production of commodity chemicals from biomass-derived

precursors that use Diels-Alder chemistry to transform furanic compounds into aromatic species of interest to the chemical industry.

These results show that despite the maturity of the field of zeolite catalysis, tantalizing new opportunities emerge from the discovery of new zeolite structures and from improvements in our control of the composition of the framework and extra-framework space.

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