

Seminar

Wednesday, April 25, 2018

11:00 AM – 206 Furnas Hall

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Professor

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On the Reaction Mechanism and the Nature of the Active Site for Standard Selective Catalytic Reduction of NO_x on Cu/SSZ-13 Zeolites

Cu/SSZ-13 zeolites are used commercially in mobile applications for the selective reduction of NO_x with ammonia. Our data suggest that the catalytic cycle is a redox one involving Cu(I) and Cu(II) species. During preparation, the Cu(II) species are the most stable and reside preferentially on six-member rings containing two aluminum sites. The Cu(II) ions first populate 2Al sites before the other remaining unpaired sites with 1Al are populated, Cu(II)OH. These sites could be counted ex situ through vibrational and X-ray absorption spectroscopies (XAS) and chemical titrations. In spite of an apparent chemical difference, the two sites had similar standard SCR reaction turnover rates, apparent activation energies and apparent reaction orders at SCR conditions, even on zeolite frameworks other than SSZ13. Operando XAS experiments showed similar Cu(I) – Cu(II) fractions at steady state, demonstrating their similarity for SCR redox chemistry. Finally, it is shown that two NH₃-solvated Cu(I) ions tethered to the zeolite framework migrate to form a dimer that mediated by O₂ is oxidized from Cu(I) to Cu(II), react with NO and break apart to close the catalytic cycle. This study also illustrates the necessity of a collaboration among experts in synthesis, characterization, kinetics and theory for a chance to understand the catalytic cycle.

Refreshments at 10:45



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