Nitrogen Oxides (NOx) are by-product pollutants of combustion reactions in air. Their selective reduction in oxygen rich environments is a key challenge in realizing more stringent emission regulations. NOx Selective Catalytic Reduction (SCR) is the catalyzed reaction through which NOx species are selectively reduced in an excess oxygen environment. Metallic SCR catalysts have problems with poisoning and selectivity, so recent focus has shifted to the use of zeolite catalysts, which are porous aluminosilicate materials. The metal-exchanged zeolite Cu-SSZ-13 in particular has shown excellent activity, selectivity, and hydrothermal stability and has already seen industrial usage. The fundamental reasons why Cu-SSZ-13 exhibits these traits are not well understood. **A primary obstacle for further improvement of SCR catalysts is a lack of molecular-level understanding of SCR activity in current catalysts**.

Our previous work has identified single Cu species in the six membered ring of Cu-SSZ-13 as an active site of SCR. Here we investigate the mechanistic chemistry through which the reaction proceeds at those sites utilizing hybrid (HSE06) Density Functional Theory (DFT) planewave calculations. To accomplish this we:

1) Probe the interaction between the SCR gas species and the active site using DFT and *ab-initio* molecular dynamics. We then validate the calculation results through comparison to *operando* X-ray adsorption experiments performed by our collaborators at Purdue and Argonne National Laboratory

2) Construct a thermodynamic phase diagram to compare the relative stability of the adsorbed species as well as other possible reaction intermediates under SCR conditions.

3) Elucidate a reaction mechanism for SCR that is consistent with the findings above.

We conclude that the SCR mechanism involves cyclic redox of the isolated Cu species between its 2+ and 1+ oxidation states. We show that no single gas species will both spontaneously adsorb and reduce/oxidize the Cu and thus the redox require formation of intermediates. The reduction half cycle occurs via an intermediate formed selectively in the presence of O_2 from NH_3 and NO. This formation generates an excess acid site, which has been quantified experimentally. The oxidation half cycle involves an intermediate formed from NO and O_2 to form nitrite, which reacts with the NH_4^+ formed from the generated acid site.

