Site-Specific Reactivity of Copper Chabazite Zeolites with Nitric Oxide, Ammonia, and Oxygen

In-situ electron paramagnetic resonance (EPR) spectroscopy was applied to dilute copper chabazite (CHA) zeolites under gas flows relevant for the selective catalytic reduction of NO with ammonia (NH3-SCR). Under both reducing and oxidizing conditions, we observed differences in reactivity between the different monomeric copper sites present: When reducing with NO+NH3, the rate of reduction of Cu2+ sites depends on NH3 coverage. The subsequent oxidation with O2 results in a clean EPR spectrum of only one type of copper site, whereas oxidation in NO+O2 gives two types of copper sites. The rate of oxidation differs significantly between reaction with O2 alone and with NO+O2 together. Thus it was revealed that [Cu(NH3)2]+ complexes, which are regarded to be only weakly associated with the framework, nevertheless have different reactivity depending on the Al distribution in the proximity. The observed differences in reactivity of copper sites has implications for the mechanistic understanding of NH3-SCR with Cu-zeolites.