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Authors
Aranifard, Sara
Bell, Alexis T
Keil, Frerich J
et al.

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Kinetic modeling of nitrous oxide decomposition on Fe-ZSM-5 in the presence of nitric oxide based on parameters obtained from first-principles calculations†‡

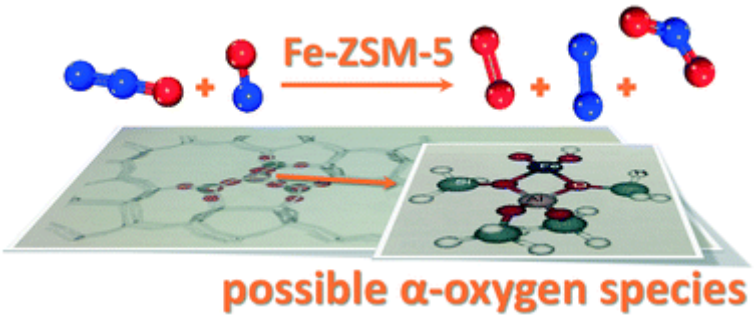
Sara Aranifard, *a Alexis T. Bell, b Frerich J. Keil c and Andreas Heyden *a

Author affiliations

Abstract

A variety of experiments for the N_2O decomposition over Fe-ZSM-5 catalysts have been simulated in the presence and absence of small amounts of nitric oxide and water vapor. A comprehensive reaction mechanism over mononuclear iron sites was considered, and all elementary reaction rate constants were obtained from density functional theory and transition state theory. Various experimental observations,

such as the acceleration of the N_2O decomposition in the presence of nitric oxide at low temperatures, can be described with the studied reaction mechanism on mononuclear iron sites. No other iron species, such as binuclear iron, are necessary for explaining experimental observations. At high temperatures, $Z^-[FeO]^+$ sites are active for N_2O decomposition, forming either $Z^-[OFeO]^+$ and $Z^-[FeO_2]^+$ sites on which a second N_2O can decompose to form $Z^-[OFeO_2]^+$ sites from which O_2 can rapidly desorb. At low temperatures, nitric oxide activates water poisoned $Z^-[Fe(OH)_2]^+$ sites to form active $Z^-[FeOH]^+$ species. The catalytic cycle on $Z^-[FeOH]^+$ involves N_2O dissociation to form $Z^-[OFeOH]^+$ sites that are inactive for a second N_2O decomposition. Instead, NO adsorbs and NO_2 desorbs in order to regenerate the $Z^-[FeOH]^+$ sites. On all active iron sites, the first N_2O dissociation step is the most critical rate-controlling step. The concentration of nitric oxide and water vapor together determine at which temperature the switch between the most dominant active site (hydroxo- *versus* oxo–iron species) occurs. Overall, this study motivates the investigation of $Z^-[OFeOH]^+$ sites as potential α-oxygen species that can oxidize various hydrocarbons at low temperatures.



Supplementary files

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