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### Title

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

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From the journal:

## Catalysis Science & Technology

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



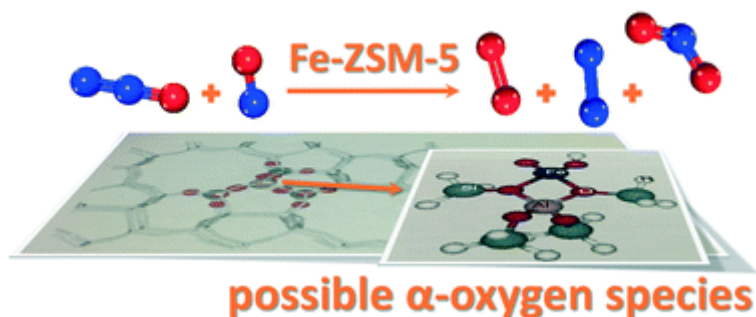
[Sara Aranifard](#), <sup>\*a</sup> [Alexis T. Bell](#), <sup>b</sup> [Frerich J. Keil](#), <sup>c</sup> and [Andreas Heyden](#), <sup>\*a</sup>

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## Abstract

A variety of experiments for the N<sub>2</sub>O 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  $\text{N}_2\text{O}$  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,  $\text{Z}^-[\text{FeO}]^+$  sites are active for  $\text{N}_2\text{O}$  decomposition, forming either  $\text{Z}^-[\text{OFeO}]^+$  and  $\text{Z}^-[\text{FeO}_2]^+$  sites on which a second  $\text{N}_2\text{O}$  can decompose to form  $\text{Z}^-[\text{OFeO}_2]^+$  sites from which  $\text{O}_2$  can rapidly desorb. At low temperatures, nitric oxide activates water poisoned  $\text{Z}^-[\text{Fe}(\text{OH})_2]^+$  sites to form active  $\text{Z}^-[\text{FeOH}]^+$  species. The catalytic cycle on  $\text{Z}^-[\text{FeOH}]^+$  involves  $\text{N}_2\text{O}$  dissociation to form  $\text{Z}^-[\text{OFeOH}]^+$  sites that are inactive for a second  $\text{N}_2\text{O}$  decomposition. Instead,  $\text{NO}$  adsorbs and  $\text{NO}_2$  desorbs in order to regenerate the  $\text{Z}^-[\text{FeOH}]^+$  sites. On all active iron sites, the first  $\text{N}_2\text{O}$  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  $\text{Z}^-[\text{OFeOH}]^+$  sites as potential  $\alpha$ -oxygen species that can oxidize various hydrocarbons at low temperatures.


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### Supplementary information

PDF (320K)

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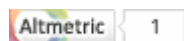
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