Insights into the mechanism of simultaneous N_2O and NO abatement over iron-zeolites by modulated excitation spectroscopy

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Nitrous oxide (N_2O) and nitric oxide (NO) are air pollutions that strongly affect human health and the environment. Scientific research on the abatement of both exhaust gas components has experienced a tremendous interest due to industrial processes and the forthcoming advent of ammonia-fueled ship engines, which will result in increasing emissions of these pollutants. Iron-exchanged zeolites proved to be suitable for either the individual abatement of N_2O and NO or the combined selective catalytic reduction (SCR) of N_2O and NO by NH_3 (N_2O-NO-SCR).[1] For the identification and monitoring of the redox processes and dynamics of adsorbed species on iron-zeolites in these processes, we combined catalytic experiments with transient spectroscopic methods using the modulated excitation (ME) approach[2] (ME-XAS, ME-EPR, ME-DRUV, ME-DRIFTS). The application of these methods allowed to develop a mechanistic understanding of the N_2O decomposition reaction over iron-zeolites and embed it into a comprehensive understanding of N_2O-NO-SCR chemistry: i) N_2O is activated on square planar isolated Fe_{β}^{2+} sites in form of Fe_{β}^{3+}-OH, ii) Fe_{β}^{2+}-NO species drives utilization of Fe_{β}^{3+}-OH, promoting iii) the Fe_{β}^{3+}-OH/ Fe_{β}^{2+} redox transition and the oxidative activation of Fe_{β}^{2+}-NO to Fe_{β}^{3+}-HONO. Finally, iv) Fe_{β}^{3+}-HONO is reduced by reactive $NH_{3,BAS}$ producing N_2 and water through NH_4NO_2 and closing the catalytic cycle.[3]

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