

The Effect of Framework Structure on N₂O Formation over Cu-Based Zeolites during NH₃-SCR Reactions

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Abstract : Nitrous oxide (N₂O), which is generally formed as a byproduct of industrial chemical processes and fossil fuel combustion, has attracted considerable attention due to its destructive role in global warming and ozone layer depletion. From various developed technologies used for lean NO_x reduction, the selective catalytic reduction (SCR) of NO_x with ammonia is presently the most applied method. Therefore, the development of catalysts for efficient lean NO_x reduction without forming N₂O in the process, or only forming it to a very small extent from the exhaust gases is of crucial significance. One type of catalysts that nowadays are used for this aim are zeolite-based catalysts. It is owing to their remarkable catalytic performance under practical reaction conditions such as high thermal stability and high N₂ selectivity. Among all zeolites, copper ion-exchanged zeolites, with CHA, MFI, and BEA framework structure (like SSZ-13, ZSM-5 and Beta, respectively), represent higher hydrothermal stability, high activity and N₂ selectivity. This work aims at investigating the effect of the zeolite framework structure on the formation of N₂O during NH₃-SCR reaction conditions over three Cu-based zeolites ranging from small-pore to large-pore framework structure. In the zeolite framework, Cu exists in two cationic forms, that can catalyze the SCR reaction by activating NO to form NO⁺ and/or surface nitrate species. The nitrate species can thereafter react with NH₃ to form another intermediate, ammonium nitrate, which seems to be one source for N₂O formation at low temperatures. The results from in situ diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS) indicate that during the NO oxidation step, mainly NO⁺ and nitrate species are formed on the surface of the catalysts. The intensity of the absorption peak attributed to NO⁺ species is higher for the Cu-CHA sample compared to the other two samples, indicating a higher stability of this species in small cages. Furthermore, upon the addition of NH₃, through the standard SCR reaction conditions, absorption peaks assigned to N-H stretching and bending vibrations are building up. At the same time, negative peaks are evolving in the O-H stretching region, indicating blocking/replacement of surface OH-groups by NH₃ and NH₄⁺. By removing NH₃ and adding NO₂ to the inlet gas composition, the peaks in the N-H stretching and bending vibration regions show a decreasing trend in intensity, with the decrease being more pronounced for increasing pore size. It can probably be owing to the higher accumulation of ammonia species in the small-pore size zeolite compared to the other two samples. Furthermore, it is worth noting that the ammonia surface species are strongly bonded to the CHA zeolite structure, which makes it more difficult to react with NO₂. To conclude, the framework structure of the zeolite seems to play an important role in the formation and reactivity of surface species relevant for the SCR process. Here we intend to discuss the connection between the zeolite structure, the surface species, and the formation of N₂O during ammonia-SCR.

Keywords : fast SCR, nitrous oxide, NO_x, standard SCR, zeolites

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