The reliability study on the Cu/CHA NH$_3$-SCR catalysts: SO$_3$ and Na ions poisoning

Chen Wang and Xue Kang
North University of China, China

The deactivation mechanism of Cu/CHA ammonia selective catalytic reduction catalysts by SO$_3$ poisoning has been systematically investigated using a range of analytical techniques. In order to study the influence of SO$_3$ poisoning on active Cu$^{2+}$ ions and the zeolite framework, different sulfate samples were prepared with different contents of SO$_3$ (0-20%) in SO$_3$ under same poisoning condition. The results reveal the NO conversion of samples poisoned by SO$_3$ decreased more than that poisoned by SO$_2$ when temperature ranged between 100° and 600°. The TPR and EPR results demonstrate that SO$_3$ poisoning does a significant influence on the amount of active Cu$^{2+}$ ions than SO$_2$ does. The kinetic results illustrate the SO$_3$ poisoning has no impact on the apparent activation energy (Ea) of NH$_3$-SCR reaction over Cu/CHA catalysts. The reason of NH$_3$-SCR activity declining is the reduction of the number of isolated Cu$^{2+}$ ions among the kinetic temperature regions. The ex-situ DRIFTS and BET results expose that the SO$_3$ poisoning could decrease the crystallization by damaging Si-OH-Al structure. The NH$_3$-SCR activity at high temperature decline because of the NH$_3$ migration difficulty resulted by structure damaging. Cu/CHA catalysts have been found to be affected by alkali and alkali earth ions; however, the poisoning mechanism is still unclear. In order to investigate Na poisoning effects and its mechanism on Cu/SAPO-34 and Cu/SSZ-13, five samples with different Na contents were synthesized. The Na effects on the structure, Cu species, and NH$_3$-SCR reaction over Cu/CHA were characterized through XRD, BET, NH$_3$-TPD, ex-DRIFTS, H$_2$-TPR, EPR, activity tests and kinetic experiments, and CO$_2$-DRIFTS were used to probe the types of Na species. The results indicate that the introduced Na$^+$ exchanged with H$^+$ and Cu$^{2+}$, and it mainly substituted H$^+$ from Si-OH-Al, then H$^+$ from surface OH, finally isolated Cu$^{2+}$. The exchanged H$^+$ led to the structure damaging of Cu/CHA by dealumination, and the exchanged Cu$^{2+}$ aggregated and formed CuOx species. The NH$_3$-SCR activity decreased with Na contents, and the loss of isolated Cu$^{2+}$ and CHA structure was responsible for the performance deactivation.

chenwang87@nuc.edu.cn