

Quantification and Redox Property of the Oxygen-Bridged Cu_2^+ Dimers as the Active Sites for the NO Decomposition over Cu-ZSM-5 Catalysts

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For a range of Cu-ZSM-5 catalysts with different Cu-exchange levels on the two kinds of ZSM-5 with different Si/Al ratios, temperature programmed reduction using CO (CO-TPR) followed by H_2 (H_2 -TPR), and temperature programmed desorption of oxygen (O_2 -TPD) were conducted using an online mass spectrometer to characterize and quantify the copper species on the catalysts in the calcined state. Copper species on the ZSM-5 were quantitatively characterized to exist as Cu^{2+} , $(\text{Cu}^{2+}-\text{O}^{2-}-\text{Cu}^{2+})^{2+}$ and CuO after calcination in oxygen environment. The N_2 formation activities of the catalysts in the decomposition of NO were well correlated with the quantified catalytic amounts of the Cu^{2+} ions involved in the Cu-dimers, $(\text{Cu}-\text{O}-\text{Cu})^{2+}$. The mol fraction of the Cu ions present as the Cu-dimers increased at the sacrifice of the isolated Cu^{2+} with increasing Cu ion exchange level. Oxygen that could be thermally desorbed from the oxidized catalysts in the O_2 -TPD was responsible for the reduction of the Cu-dimers. It was concluded that the decomposition of NO over Cu-ZSM-5 catalyst proceeded by the redox of Cu-dimer species, $(\text{Cu}-\text{O}-\text{Cu})^{2+}$, as active centers.