Ti or Sn doping as a way to increase activity and sulfur tolerance of Mn/CeO$_2$ catalyst for low temperature NH$_3$ selective catalytic reduction of NO

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Abstract Text

Mn/CeO$_2$ catalysts modified by doping of Ti or Sn were investigated for low temperature selective catalytic reduction (SCR) of NO by NH$_3$ with the aim of studying the effects of Ti, Sn doping on the catalytic performance. Ceria-based solid solutions (Ce0.8Ti0.2O$_2$ and Ce0.8Sn0.2O$_2$) were synthesized via inverse co-precipitation, and used as supports to prepare MnOx/Ce0.8M0.2O$_2$ (M =Ti$^{4+}$, Sn$^{4+}$) catalysts through wetness impregnation method. The results showed that doping of Ti or Sn to the CeO$_2$ support increase the NO removal efficiency. A NO conversion of more than 90 % was obtained over the Mn/CeTi catalyst at the temperature window of 175 $\sim$ 300 $^\circ$C under a gas hourly space velocity (GHSV) of 60,000 mL·g$^{-1}$·h$^{-1}$. Catalysts modified by Ti and Sn were also found to obtain higher SO$_2$ resistance than Mn/CeO$_2$ catalyst. More than 90% NO conversion and 95% N$_2$ selectivity could be provided by Mn/CeTi catalyst in the presence of 100 ppm SO$_2$ at 250 $^\circ$C for 10 h. A series of characterization techniques, namely XRD, BET, H$_2$-TPR, XPS, NH$_3$-TPD and in situ DRIFTS were used to elucidate the structure and surface properties of the obtained supports and catalysts. The results indicate that doping of Ti or Sn brings about catalysts with favorable properties such as higher BET surface area, better oxygen storage capacity and stronger surface acidity. The relative amount of Mn$^{4+}$, Ce$^{3+}$, adsorbed oxygen species and oxygen vacancies on the surface of catalysts are in the order of Mn/CeTi$^{[U+FF1E]}$ Mn/CeSn$^{[U+FF1E]}$ Mn/CeO$_2$, which is thought to make positive a contribution to the low-temperature SCR activity. The promoted SCR activity is considered as well to be related to the dual redox cycles in Mn/CeTi (Mn$^{4+}$ + Ce$^{3+}$ $\leftrightarrow$ Mn$^{3+}$ + Ce$^{4+}$, Ce$^{4+}$ + Ti$^{3+}$ $\leftrightarrow$ Ce$^{3+}$ + Ti$^{4+}$) and Mn/CeSn (Mn$^{4+}$ + Ce$^{3+}$ $\leftrightarrow$ Mn$^{3+}$ + Ce$^{4+}$, Ce$^{4+}$ + Sn$^{2+}$ $\leftrightarrow$ Mn$^{3+}$ + Ce$^{4+}$ + Sn$^{4+}$) catalysts.