

Effects of Mercury on SCR-catalysts

Michael J.T. Hocquel, Presenter

Email: hocquel@ivd.uni-stuttgart.de ; Telephone : (+49)-711-685-7759 ; Fax : (+49)-711-685-3491

Sven Unterberger, Co-author

Email: unterberger@ivd.uni-stuttgart.de ; Telephone : (+49)-711-685-3572 ; Fax : (+49)-711-685-3491

Klaus R.G. Hein, Co-author

Email: hein@ivd.uni-stuttgart.de ; Telephone : (+49)-711-685-3487 ; Fax : (+49)-711-685-3781

Institute of Process Engineering and Power Plant Technology, Pfaffenwaldring 23, 70569 Stuttgart, Germany

Summary

SCR-DeNOx-catalysts in high-dust applications have been reported to positively influence the oxidation of mercury. This assumption is based on results reported from measurements at plants firing hard coals with Cl-contents in the range of 0,1 mass-%. At low-dust applications and/or at plants with low chlorine coals less effective oxidation and sometimes bad mass balance closures have been reported. The role of SCR-DeNOx-catalysts and other fly ash components (i.e. CaO) within the mercury oxidation process is critical discussed in the following.

In several experimental studies the effect of SCR-DeNOx-catalyst on the behavior of mercury has been investigated at the Institute of Process Engineering and Power Plant Technology. Results from laboratory tests with different honeycomb catalysts (low- and high-dust type, new and regenerated) and different synthetic gas compositions of HCl, H₂O, SO₂, O₂, Hg_{total}, N₂ indicated different oxidation rates for mercury. Temperature has been varied between 230 and 340°C with HCl-concentrations between less than 1 mg/m³ and 150 mg/m³. Hg-CEM measurements determine mercury species with and without the SCR-DeNOx-catalytic converter.

Results show a strong influence of both temperature and HCl-concentration on the behavior of mercury and the ongoing mechanisms. A significant conversion of Hg⁰ to HgCl₂ with sufficient high HCl concentrations has been observed. Within the investigated temperature range and HCl-concentrations of > 60 mg/m³ more than 90 % of mercury is measured as HgCl₂ after the catalyst. Lower HCl-concentrations result in an incomplete oxidation. For very low concentrations a reductive effect of the catalyst has been observed resulting in higher elemental mercury concentrations compared with values without the catalyst. Tests with single powdery catalyst components prove this observation. Furthermore CaO as an important fly ash component has proved to strongly affect the speciation of mercury at temperatures that are relevant for catalyst application. The presence of high amounts of CaO promotes higher shares of elemental mercury.

For low HCl-concentrations (< 10 mg/m³) a distinct adsorption of mercury at the catalyst has been conspicuously identified. This adsorption effect is less distinct for higher HCl-concentrations and is negligible for high HCl-concentrations (> 80 mg/m³). Through an increase of the HCl-concentration adsorbed mercury is almost completely desorbed. The reversibility of this sorption phenomenon and the curve shape of sorption and desorption is believed to be an indication for a mono-layer sorption of mercury at the active surface of the catalyst. Results show that mercury input- and output concentration at the catalytic unit could not be correlated for low HCl-concentrations because of the observed dynamic behavior of mercury. The industrial relevance of this observation has been recently demonstrated at a municipal waste incineration plant equipped with an upstream spray dryer, an activated carbon injection ahead of a baghouse, and a tail-end SCR-DeNOx catalyst. Despite the proved highly effective mercury removal unit sporadic mercury peaks have been recorded by the installed emission control equipment (Hg-CEM). A comparative analysis of the boundary conditions at the waste incineration plant with the results from laboratory experiments as well as a detailed analysis of the correlation between HCl-concentration and mercury peak has indicated that these peaks result from a spontaneous release of mercury from the catalyst initiated by an increase of HCl-concentration through Cl-peak concentrations in the fuel or/and a short-term breakdown of the spray dryer.