Mercury Oxidation over Selective Catalytic Reduction (SCR) Catalysts - Ph.d. thesis Karin Madsen - DTU Orbit (18/01/2019)

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The vanadium-based SCR catalyst used for NOx-control promotes the oxidation of elemental mercury Hg⁰ to Hg²⁺ in flue gases from coal-fired power plants. Hg²⁺ is water soluble and can effectively be captured in a wet scrubber. This means that the combination of an SCR with a wet FGD can offer an effective control option for mercury.

Laboratory experiments have been carried out to quantify the Hg⁰ oxidation that can be achieved over commercial SCR catalysts for different gas compositions, operating conditions and catalyst types. The following three net reactions have been identified as relevant for the mercury chemistry over the SCR:

R1. 2 HCl + Hg⁰ + 1/2 O₂ ↔ HgCl₂ + H₂O
R2. 2 NH₃ + 3 HgCl₂ ↔ N₂ + 3 Hg⁰ + 6 HCl
R3. 2 NO + 2 NH₃ + 1/2 O₂ ↔ 2 N₂ + 3 H₂O

where reaction R1 is the oxidation of Hg⁰ by HCl, reaction R2 is the reduction of HgCl₂ by NH₃ and reaction R3 is the DeNOx reaction.

The importance of each reaction on the achievable Hg⁰ oxidation depends on the SCR operating temperature. At T>325°C, the reduction of HgCl₂ will take place when NH₃ is present. The overall Hg⁰ oxidation will then reflect the relative rate of the Hg⁰ oxidation via reaction R1 and the HgCl₂ reduction via reaction R2. For T=250-375°C, the DeNOx reaction will inhibit the kinetics of reaction R1 by consuming active Lewis sites that must be oxidized to regain activity for Hg⁰ oxidation.

The experimental data obtained in this study indicate that vanadia Lewis sites on SCR catalysts are active in the catalytic Hg⁰ oxidation - possibly as Hg⁰ adsorption sites.

A kinetic model for the steady-state Hg⁰ oxidation over monolithic SCR reactors has been developed taking both external mass transfer, pore diffusion and reaction on the catalyst wall into account. The mercury chemistry that has been identified and quantified in the experimental investigations is incorporated in the model.

The resulting model successfully reproduces the variations in Hg⁰ oxidation over the SCR that have been experimentally observed for different gas compositions and testing conditions. This verifies that the relevant mercury chemistry has been taken into account in order to describe the catalytic Hg⁰ oxidation in a simulated flue gas. The validity of the model for describing the mercury chemistry over SCR catalysts in real flue gases is yet to be explored.

Model predictions suggest that the kinetics of the Hg⁰ oxidation over high dust SCR reactors is greatly limited by external mass transfer in the entire SCR operating temperature window if HCl≥13 ppm. For lower HCl concentrations, the surface reactivity of the SCR catalyst towards Hg⁰ oxidation can become limiting at the higher operating temperatures T>350°C, because the rate of HgCl₂ reduction will be considerable. A higher V₂O₅ load on the SCR catalyst will dampen this effect.

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