Particle Deposition in Monolithic Catalysts

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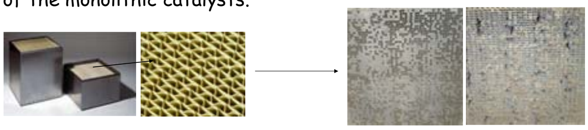
Background

The energy need and consumption of today's industrialized world have created major environmental problems such as emission of sulphur and nitrogen oxides, as well as submicron and micrometer particles. A major contributor to the emission of nitrogen oxides is combustion of coal in stationary power stations. The most commonly used method for reducing NOx in the flue gas from coal fired power stations is Selective Catalytic Reduction (SCR) of NOx – illustrated in Figure 1. For an SCR catalyst the high dust position between the economiser and air pre-heater is the most commonly used configuration. The catalyst may therefore lose activity due to chemical deactivation and physical plugging and erosion, which results in a shortened lifetime. In general submicron particles are responsible for deactivation of the catalysts by chemical poisoning, while larger particles are responsible for plugging of the channels. Particle deposition in SCR catalysts has therefore been investigated in order to understand the fundamental transport mechanisms with the purpose of being able to develop a model for plugging of the monolithic catalysts.

Example of plugging in an SCR catalyst:



The experimental investigations of particle deposition in monolithic catalysts have been carried out by using pilot and laboratory setup. Particle deposition due to fundamental mechanisms (e.g. gravity, diffusion, Saffman, electrostatic forces and turbulence) has been modelled using CFD and empirical expression.

In the present work, fundamental studies of the importance of space charge and secondary flow on the deposition of submicron particles have been carried out.

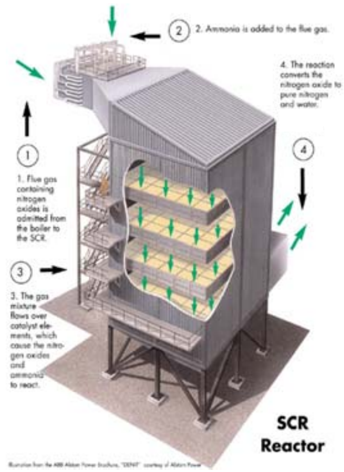


Figure 1: SCR reactor.

Experimental Setup

The experimental setup consisted of a Six-Jet Atomizer Model 9306A *TSI Inc.*, a Diffusion Dryer Model 3062 *TSI Inc.*, an Aerosol Neutralizer Model 3054 *TSI Inc.*, a 3m long (bent in about 0.75m sections) electro polished stainless steel tube for deposition of aerosol particles and two sampling lines for Scanning Mobility Particle Sizer (SMPS) measurements. Figure 2 illustrates the setup.

The flow rate in the deposition pipe was about 280 ml/min. The Scanning Mobility Particle Sizer (SMPS) system consisted of a Condensation Particle Counter (CPC) model 3775 *TSI Inc.*, and a Long Differential Mobility Analyzer (LDMA) model 3081 or a Nano Differential Mobility Analyser (NDMA) model 3085 *TSI Inc.*

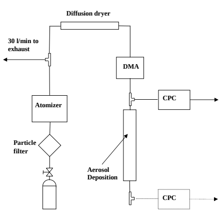


Figure 2: Experimental setup for measuring deposition of submicron particles in a 3m long (bent in about 0.75m sections) electro polished pipe with a diameter of 6mm.

CFD Modelling Brownian diffusion and electrostatic dispersion

Deposition of submicron particles in the size range between 3 nm and 1 μm in laminar pipe flow, due to Brownian diffusion has been simulated using Fluent 6.3. Additionally, electrostatic dispersion and deposition due to space charge of the submicron particles have been modelled using User Defined Scalars (UDS) in Fluent and thereby taking deposition of charged aerosol particles into account. This has been done by solving transport equations for the particle number concentration and the Poisson equation for the electrostatic potential due to space charging (induced by the charged aerosol particles) together with the Navier-Stokes equation for laminar air flow at room temperature.

The transport equation for the particle number concentration including electrostatic forces is given as follows:

$$\frac{\partial n}{\partial t} + \vec{\nabla} \cdot \left(\vec{v} n + Z n \vec{E} \right) = D \vec{\nabla}^2 n$$

where *n* is the particle number concentration, *v* the velocity field, *Z* the electrical mobility of the particles, *E* the electrical field strength and *D* the particle diffusion coefficient. The transport equation for the potential is given as:

$$\vec{\nabla}^2 \Phi = -\frac{i e n}{\varepsilon_0}$$

where *q=ie* is the total charge carried by a particle.

Figure 3: CFD model of the 3m bent pipe.

The CFD model in Figure 3 consisted of a grid with 855540 cells. Discretization was modelled using second-order schemes. Flow was laminar and incompressible.

Results and discussion

Comparison of CFD simulations for neutral (zero charge) KCl particles with experimental data for deposition of particles with a Boltzmann equilibrium charge distribution and low initial concentration, n_0 is shown in Figure 4. The figure also shows the importance of secondary flow in a bent pipe compared to a straight pipe without secondary flow, because the particle deposition is enhanced due to secondary flow which is superimposed on the main flow. The reason for this is that the secondary flow transports particles from the core of the pipe to the near-wall region where it is deposited due to diffusion.

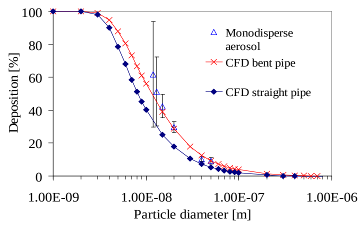


Figure 4: Deposition efficiency for charged KCl particles with a Boltzmann equilibrium charge distribution and CFD simulation of neutral particles (zero charge). Initial concentration, n_0=4-40000 [#/cm³].

Figure 5 shows CFD simulations of neutral KCl particles compared with experimental data for deposition of high concentrations of particles with e.g. a Boltzmann equilibrium charge distribution and KCl particles as delivered by the atomizer thereby carrying a high charge due to the atomization process. The initial concentration of the KCl particles was, n_0=1.7e6 [#/cm³]. Higher deposition is seen for the highly charged particles due to an additional electrostatic dispersion induced by the space charging compared to a Boltzmann equilibrium charge distribution. Figure 5 also shows a comparison of CFD simulation for unipolar particles carrying an average of 2 elementary charges.

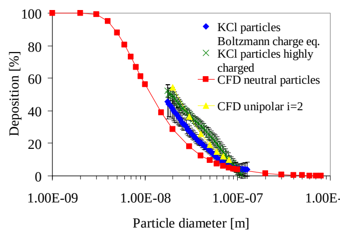


Figure 5: Deposition efficiency for charged and discharged KCl particles including CFD simulation of neutral particles. Initial concentration, n_0=1.7e6 [#/cm³].

Conclusion

- Secondary flow in a bent pipe enhances the particle deposition by transporting particles from the core of the flow to the near-wall region and thereby enhancing deposition due to Brownian diffusion.
- For high concentration of particles higher deposition rates are seen for highly charged particles compared to particles carrying a Boltzmann equilibrium charge distribution and compared to neutral particles.
- The effect of space charging is reduced for highly charged particles by lowering the particle number concentration.
- CFD has been shown to be a valuable tool for quantifying the deposition of submicron particles due to Brownian diffusion and electrostatic dispersion.

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