

# **REINHOLD ENVIRONMENTAL Ltd.**



## **2011 NO<sub>x</sub>-Combustion Round Table & Expo Presentation**

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# Dealing with fly ash deposition in SCR DeNO<sub>x</sub> – in theory and practice

Reinhold 2011 NO<sub>x</sub>-Combustion Round Table

RESEARCH | TECHNOLOGY | CATALYSTS

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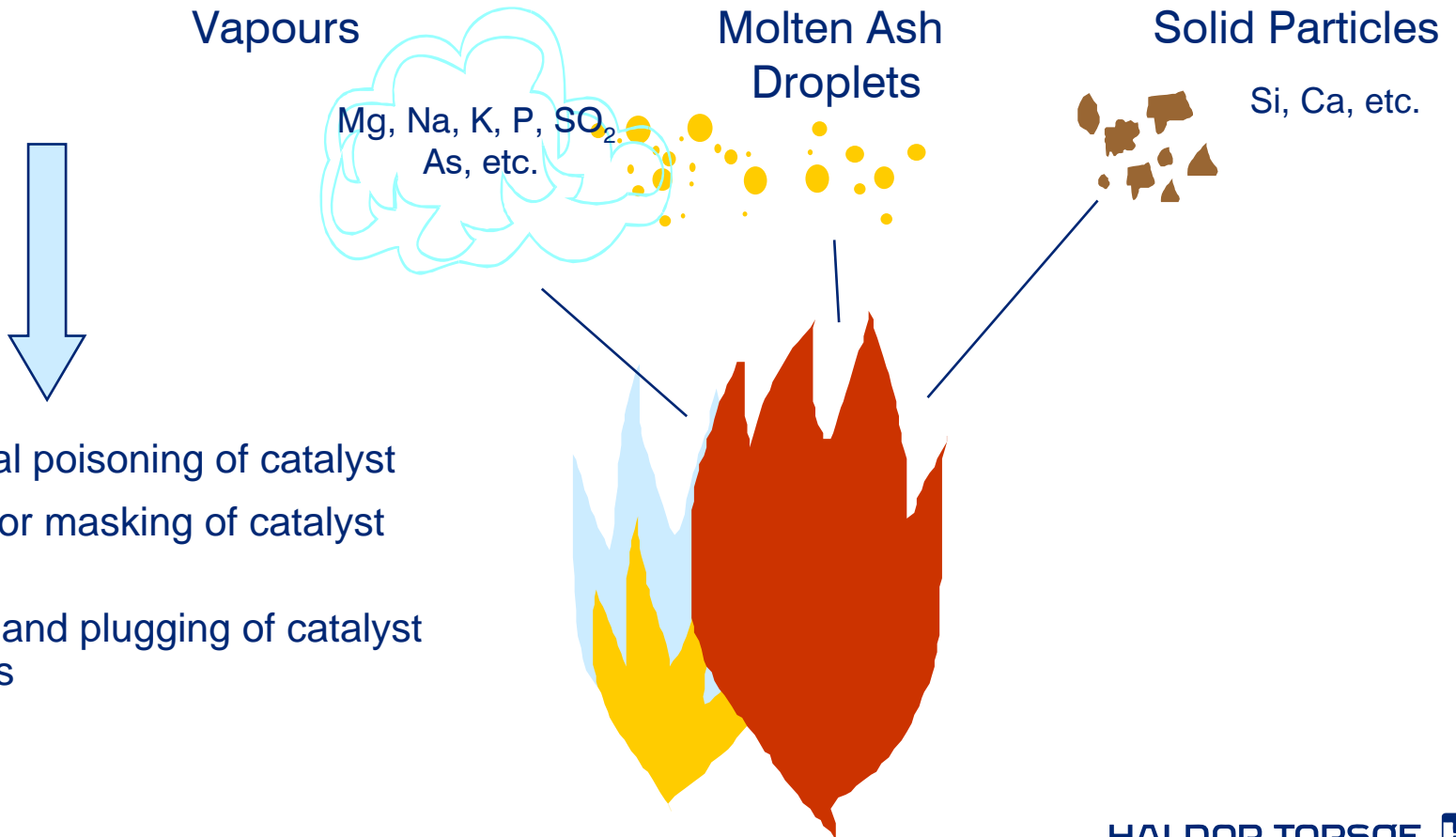
# Outline

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- Background
- PhD project – Particle dynamics in monolithic catalysts
  - Experimental work on dust deposition (LD)
  - CFD simulation of dust deposition (LD)
- Fly ash deposition in Low-Dust SCR DeNOx
  - Industrial experience
  - Short excursion to High-Dust challenges
- Summary and conclusions

# Background

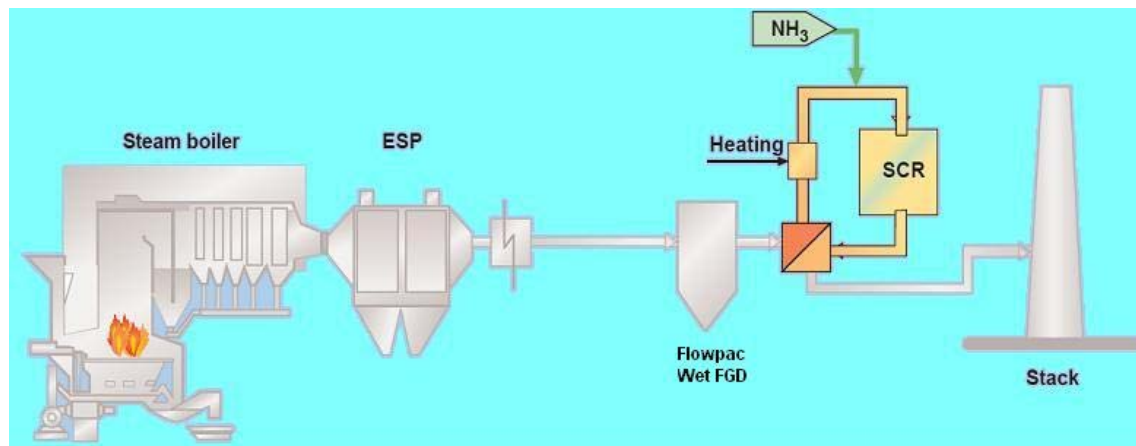
## ■ Impact of fly ash on SCR catalyst



- Chemical poisoning of catalyst
- Fouling or masking of catalyst surface
- Erosion and plugging of catalyst channels

# Background

- A major problem using SCR DeNO<sub>x</sub> is physical deactivation due to deposition of fly ash particles on the monolith
- High-dust SCR DeNO<sub>x</sub> → directly after boiler and before electrostatic precipitator (ESP)
- **Low-dust SCR DeNO<sub>x</sub>** → after ESP, or tail-end after ESP and FGD



# Background

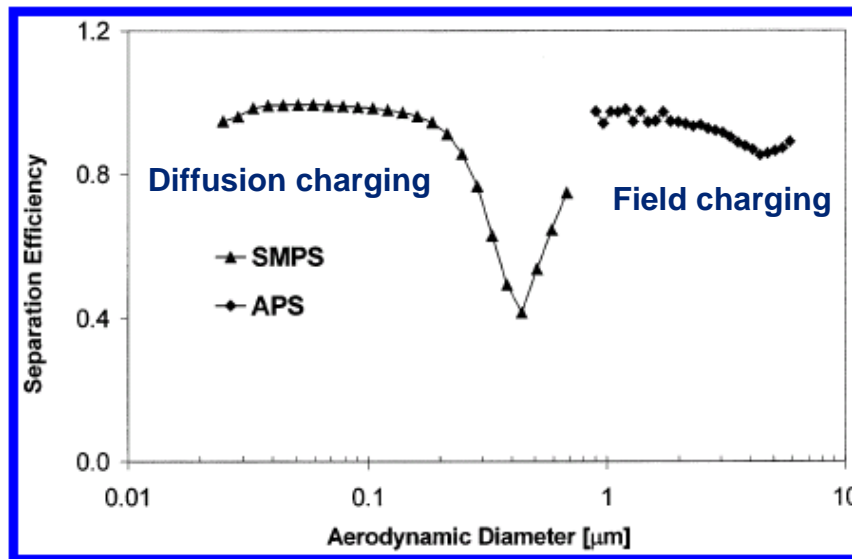
## ■ Low-dust SCR DeNO<sub>x</sub>



- Concentration of fly ash particles usually up to a few hundred mg/Nm<sup>3</sup>
- Particle sizes mainly in the range of 0.1 to a few  $\mu\text{m}$
- No soot blower and sonic horn
- 'Volcano' type of deposition pattern

# Background

- Low-dust particle size distribution as result of ESP separation characteristics
- ESP penetration window in the submicrometer size range





# PhD project 'Particle Dynamics in Monolithic Catalysts'

Michael Lykke Heiredal

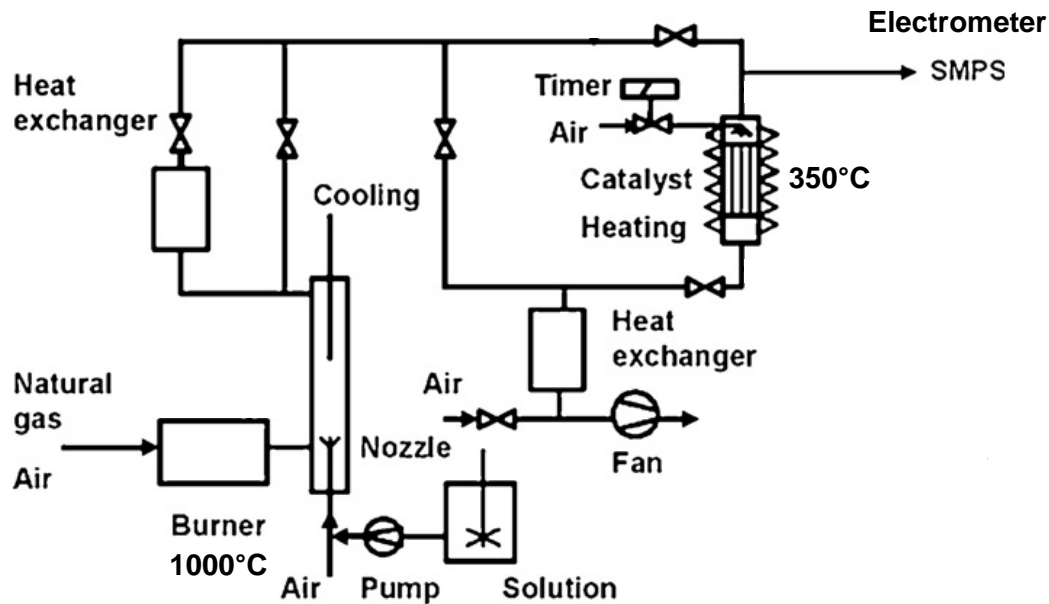
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# Objective of Ph.D. project

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- Main objective has been development of general CFD model for prediction of particle deposition and deposit build-up in Low-dust SCR DeNOx monoliths
- Main focus:
  - Pilot scale plugging experiments of SCR DeNOx monoliths under low-dust conditions
  - CFD modelling of particle deposition
  - Elucidating deposition mechanism under low-dust conditions

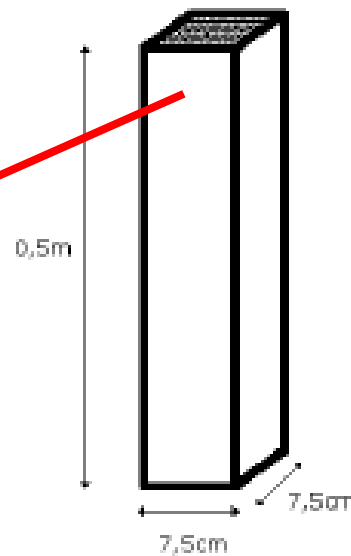
# Pilot scale experiments - setup



## Accelerated dust deposition

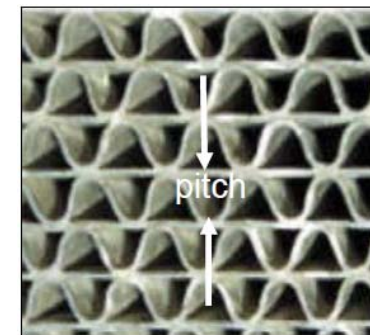
- System pressure: 5 – 30 mbar below atm. Pressure
- Volume flow: 40 Nm<sup>3</sup>/h
- Superficial velocity: 2 Nm/s
- KCl solution: 7.4 g/l
- Particle flow: 2.96 g/h
- Particle concentration in the flue gas: approx. 20 mg/Nm<sup>3</sup>
- Particle size: 0.037 – 10 μm
- **No mechanical or acoustic cleaning** (soot blower, sonic horn...)
- Negligible electrostatic forces – average charge about 0.001.

# Pilot scale experiments - setup



## TOPSØE DNX-x30

- Number of channels: ~400
- Hydraulic diameter: 3.4 mm
- Height of monolith: 0.5 m
- Length of monolith: 7.5 cm
- Width of monolith: 7.5 cm
- Catalyst open area: 0.76

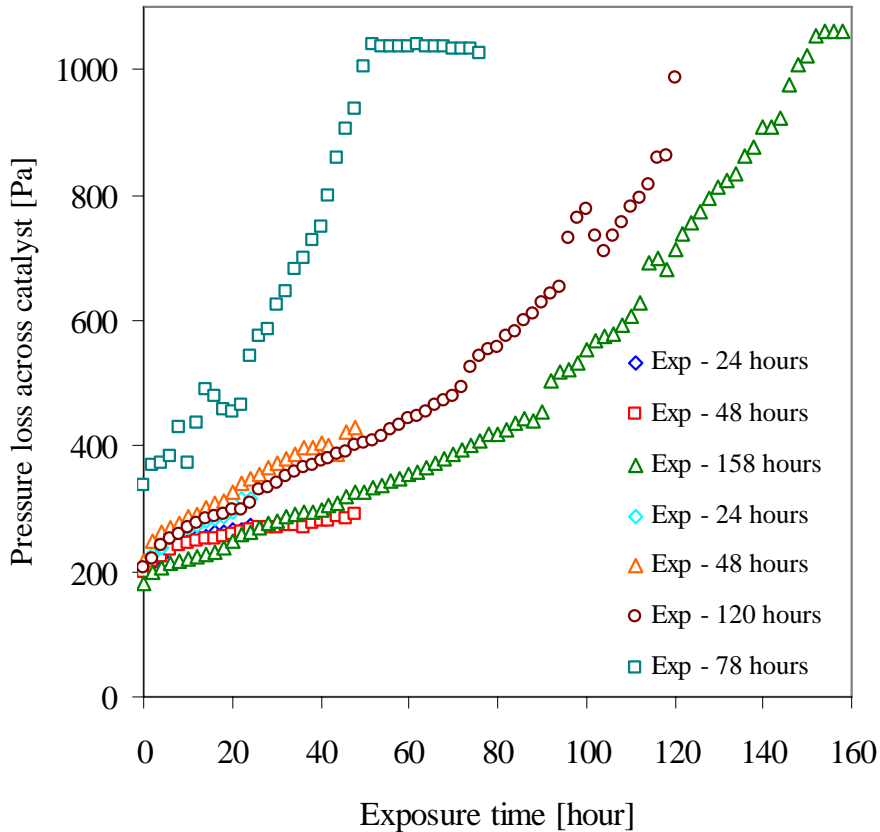


# Experimental measurements

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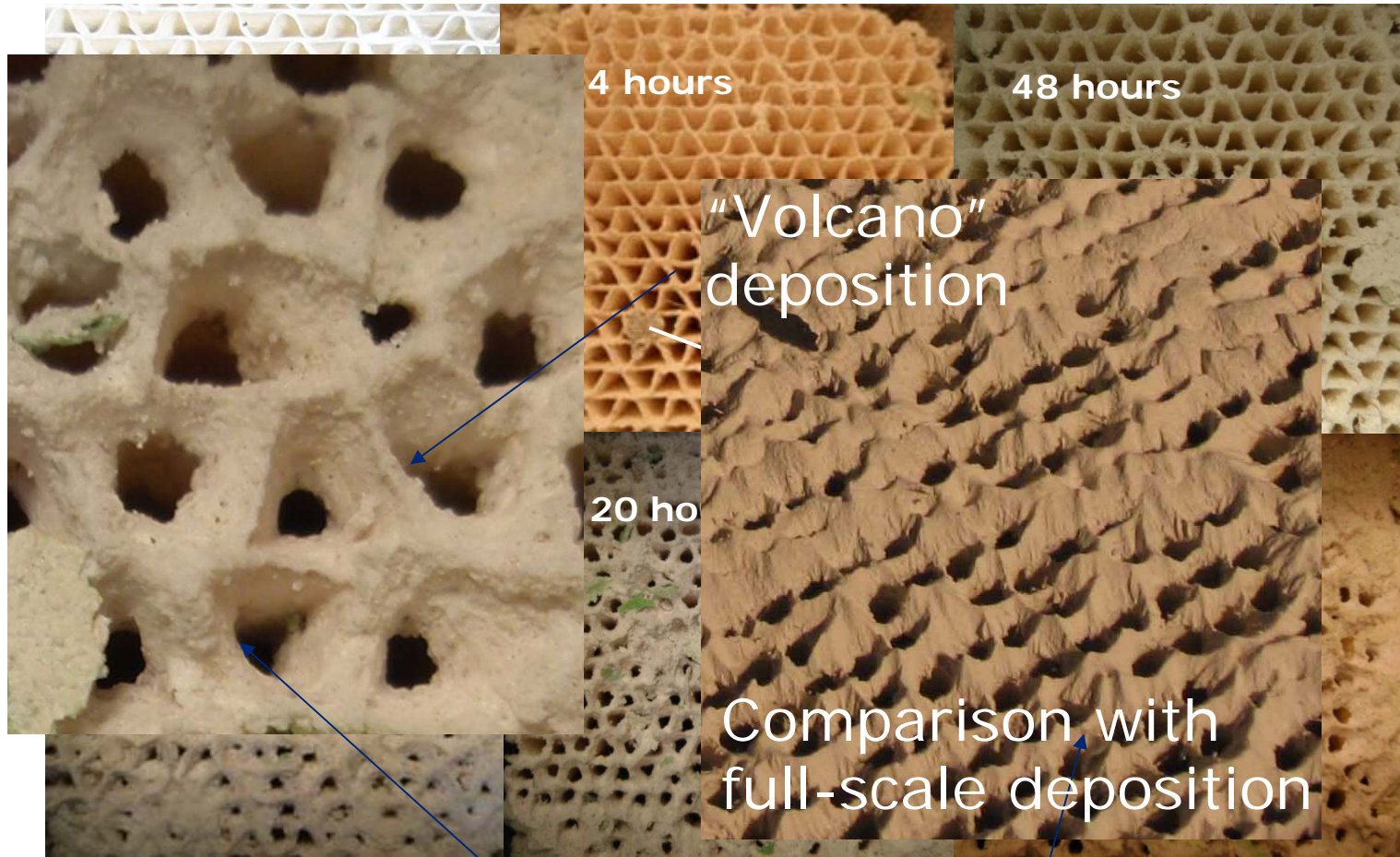
- 6 catalyst elements, DNX-x30, exposed respectively to 24, 48, 78, 120, 158 hours of operation
- Pressure rise across catalysts followed as function of exposure time
- Particle mass deposited in catalyst elements investigated by washing elements in 4-5 l ultra pure water
- Deposition profile in axial length investigated
- SEM analysis
- Average charge on fly ash particles measured

# Pressure drop across monoliths



- Reproducibility of pressure drop versus exposure time reasonable
- Second order effects observed
- The 78 hours experiment outside the trend due to new heat exchanger
- For 78 hours and 158 hours a maximum pressure is reached at about 1050 Pa due maximum under pressure available in the pilot plant

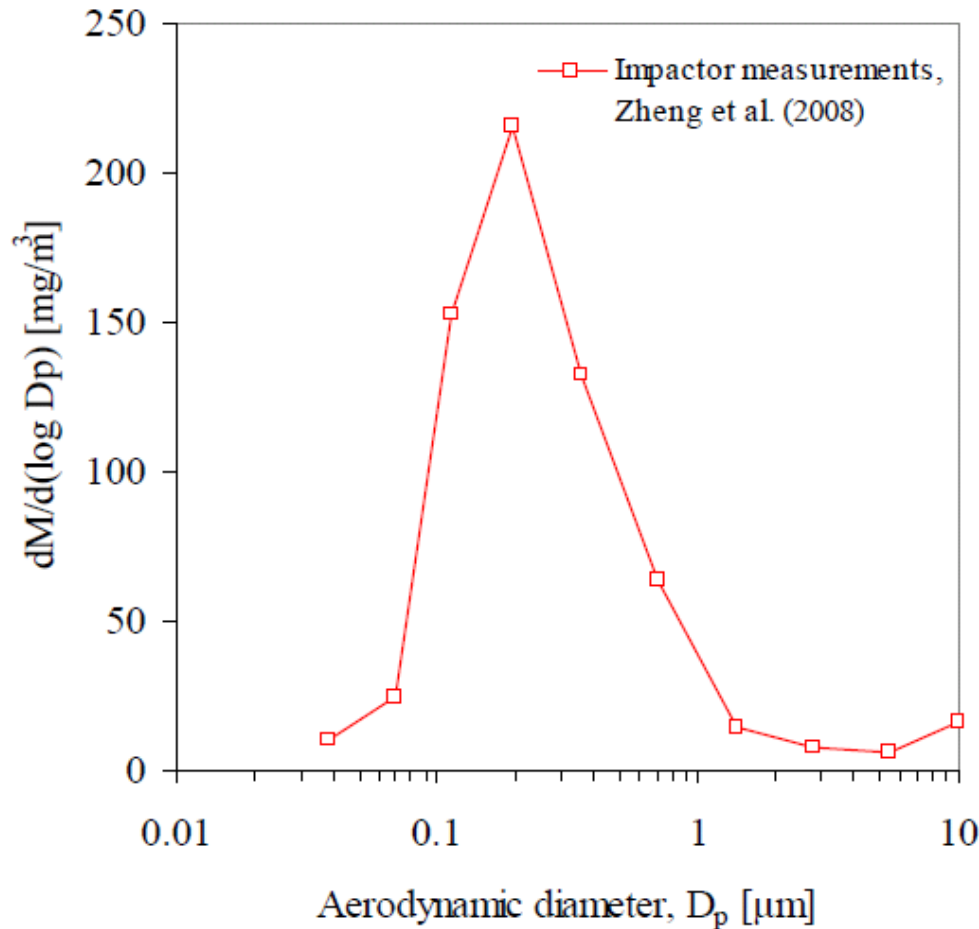
# Pilot scale plugging



"Vulcano" type of deposition

Optimizing to cylindrical channels

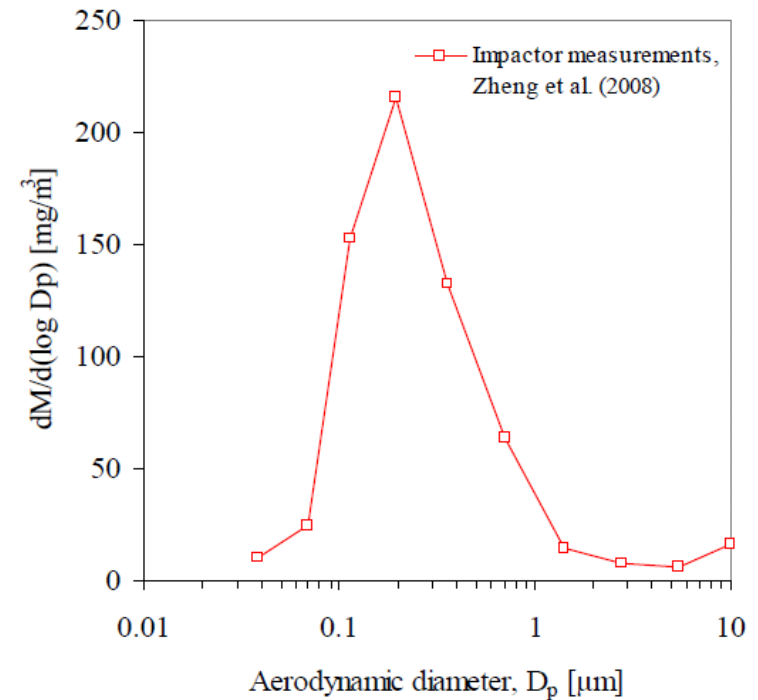
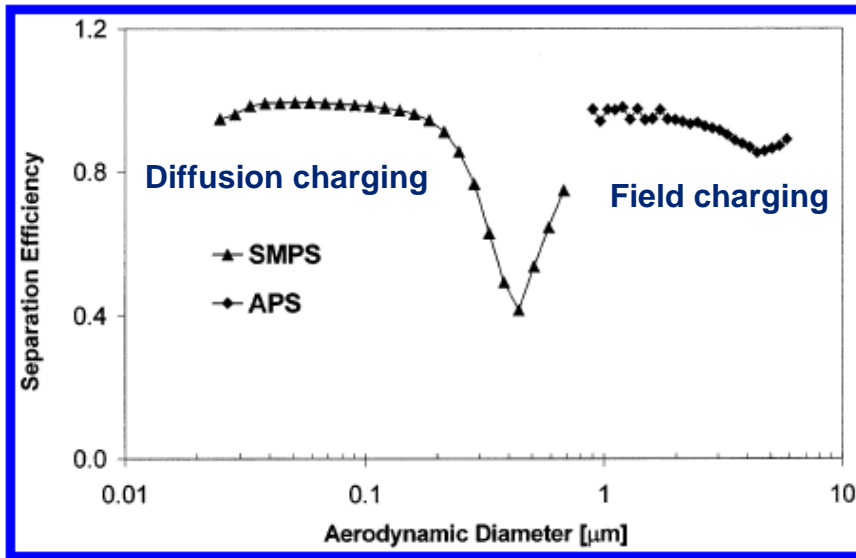
# Deposit characterization



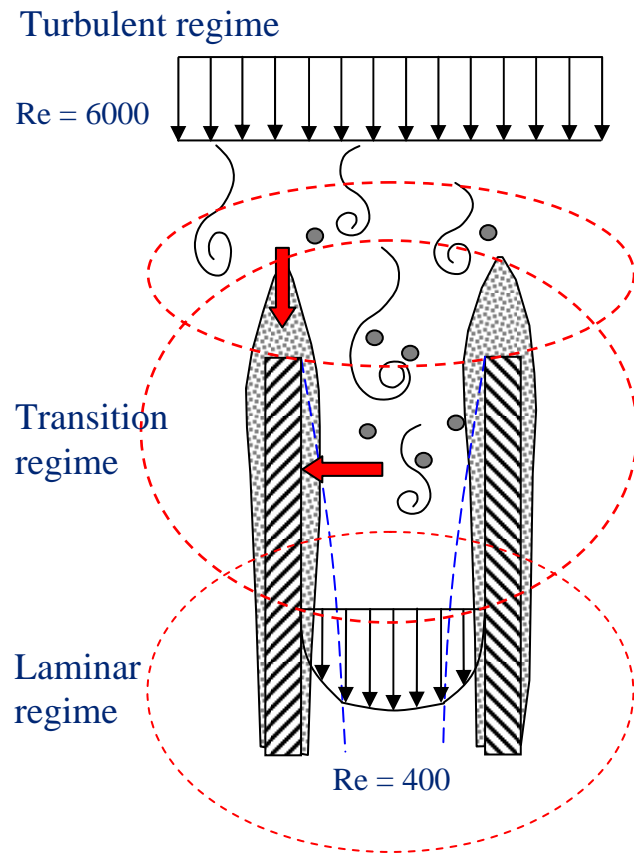
- SEM image of particles after 120 hours 30 – 40 cm downstream in the axial length of the monolith
- Consisting of agglomerates of primary particles
- Particle size in the order of 1  $\mu\text{m}$
- Impactor measurement confirms particle mainly between 0.1 and 1  $\mu\text{m}$  (submicron)

# Particle size distribution of model dust

- Realistic particle size distribution according to ESP penetration window

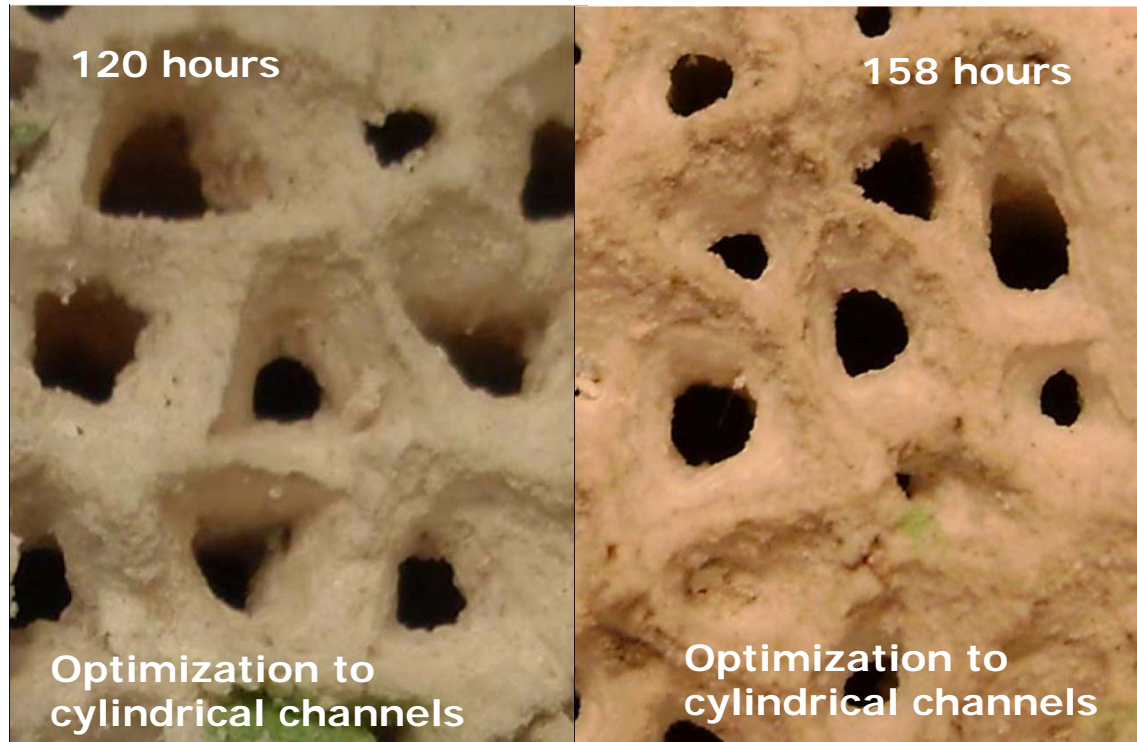


# Deposition mechanisms



- Initial dominance (short exposure time): turbulent region with inertial impaction and gravitational settling.
- Longer exposure creates streamlining and Brownian and turbulent diffusion starts to dominate
- Transition region dominated by turbulent diffusion due to turbulence convected into the monolith channels
- Laminar region dominated by Brownian diffusion (submicrometer particles) and Saffman lift (micrometer particles)

# Inlet shape optimization by deposit



- Optimization of flow geometry to cylindrical channels
- Reduced pressure loss by filling up corners with deposit
- Minimum surface area with respect to volume
- Minimized friction for certain volume flow

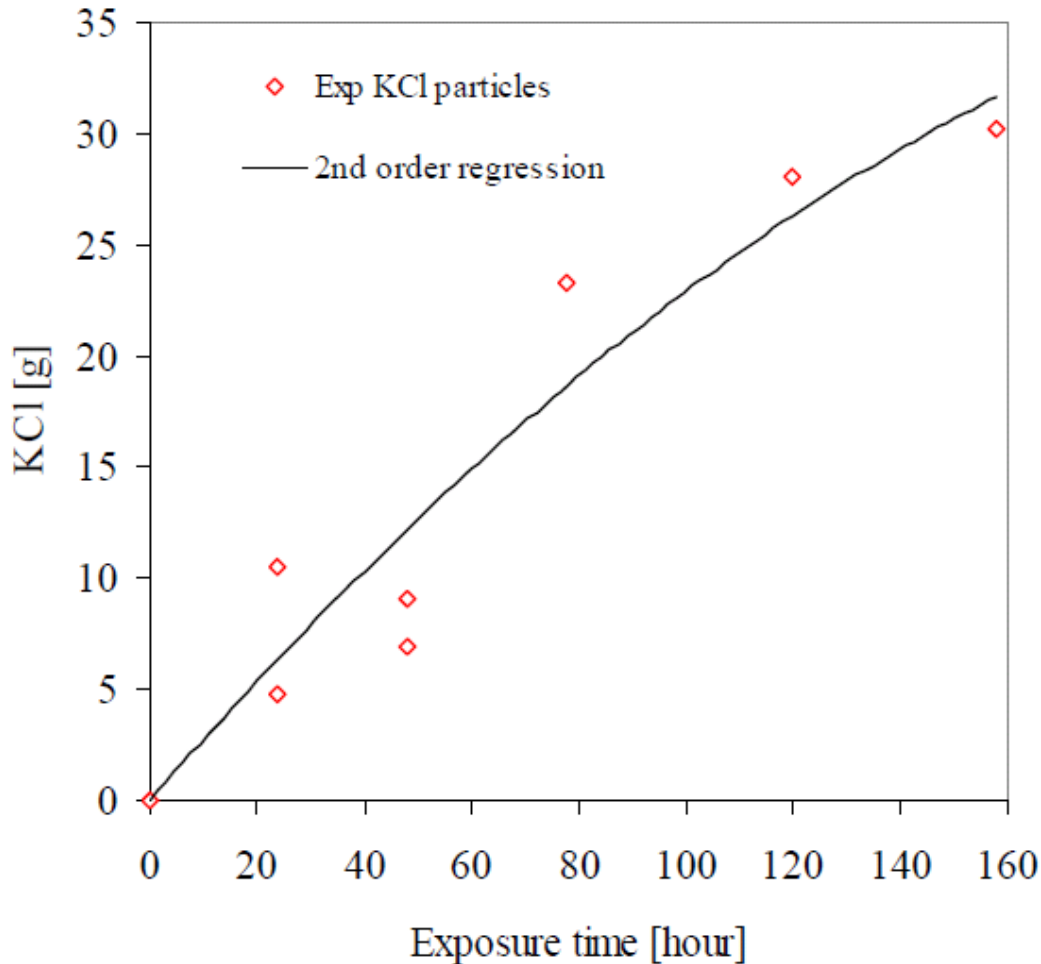
# Qualitative discussion of particle deposition

- First 0 – 48 hours of deposition no visual change in flow geometry
  - Deposition probably due to inertial impaction on the top and turbulent diffusion in the channels
  - Deposition rate constant in time and deposit does not couple back on the flow
- For longer exposure to submicron particles (above 48 hours) the inlet becomes streamlined
  - The stagnation area on top of the monolith diminishes
  - Sharp corners in the channels diminishes
  - Channel diameter in the inlet section becomes smaller

# Qualitative discussion of particle deposition

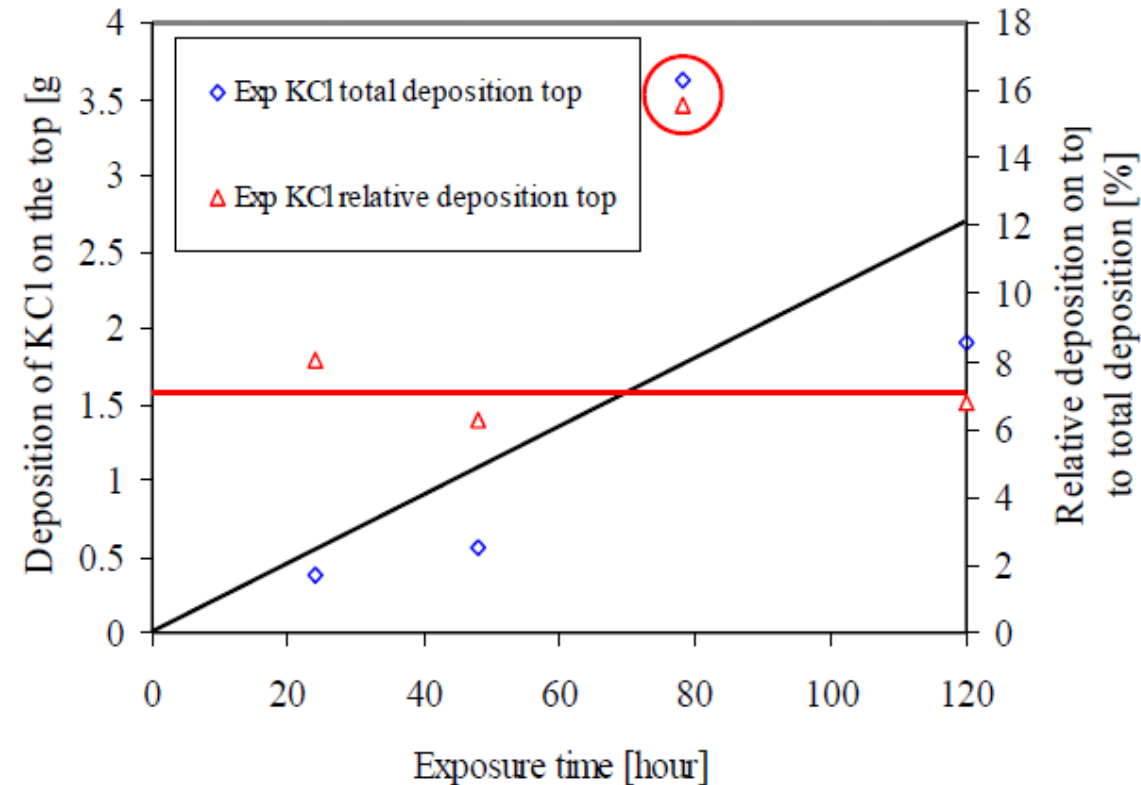
- For longer exposure
  - Deposit couples back on the flow and increases the Reynolds number in the top part of the monolith and the deposition rate increases
  - Deposition rate due to turbulent diffusion in the entrance length is expected to increase
  - Deposition rate due to inertial impaction in the flow stagnation area on top is expected to decrease
  - Deposition become more controlled by Brownian and turbulent diffusion
  - Surface roughness is expected to increase which increases the deposition rate
  - Overall increasing deposition on top and in the top part creates 'volcano' type of deposition

# Mass deposition over time



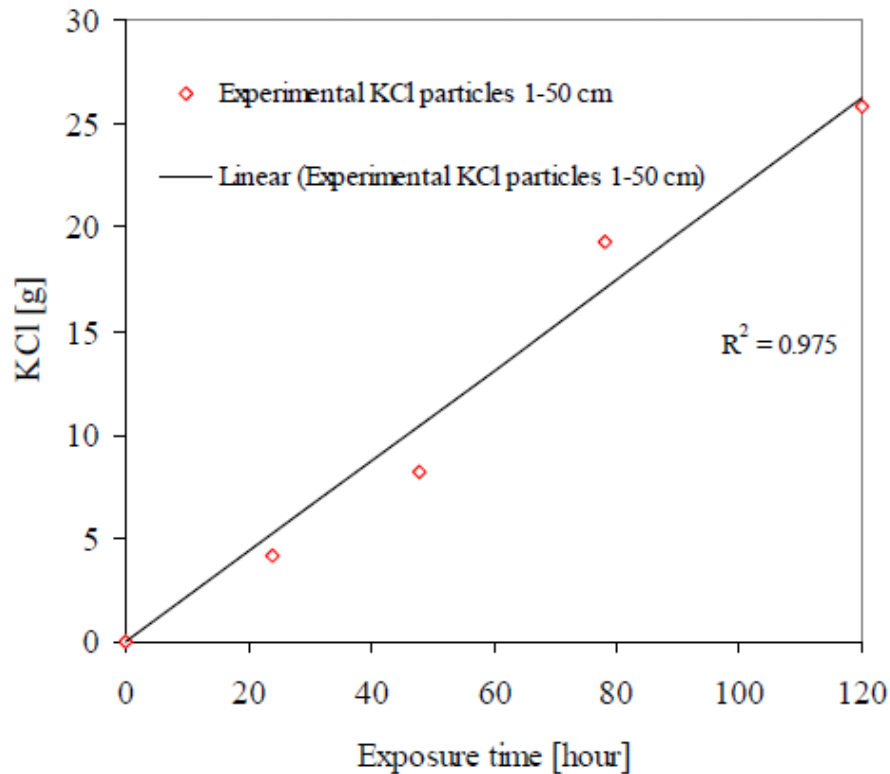
- Deposit mass after 158 hours approx. 30.3 g
- After 48 to 78 hours data indicate decreasing deposition rate

# Mass deposition on top of the channels



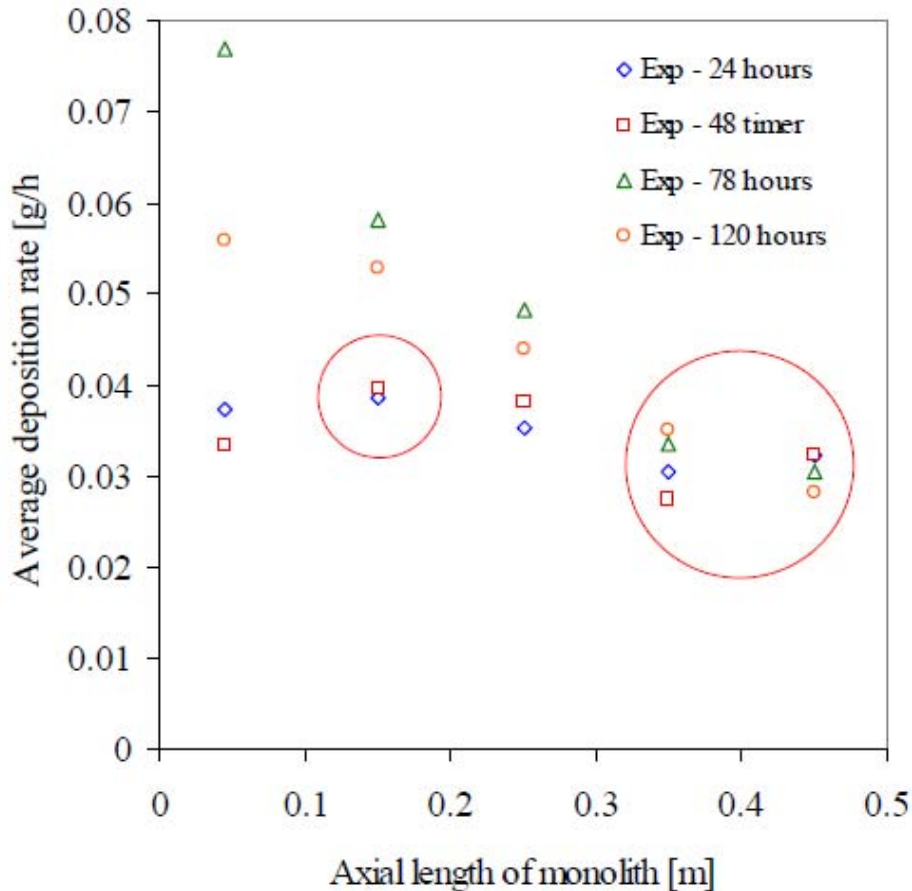
- Deposit on top of the monolith – first 0-1 cm
- Data show increasing trend (linear build-up) in mass deposition
- Relative deposition is rather constant at 6-7 %
- Data point at 78 hours outside range

# Mass deposition in the channels



- Deposit in the channels – 1-50 cm
- Data show linear build-up over time
- No data for 158 hours

# Average mass deposition rate



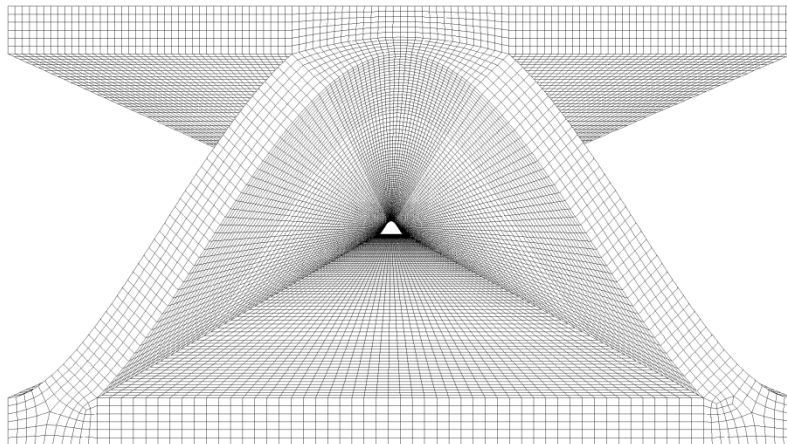
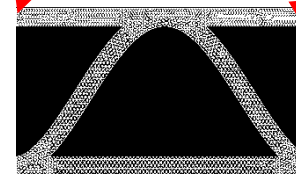
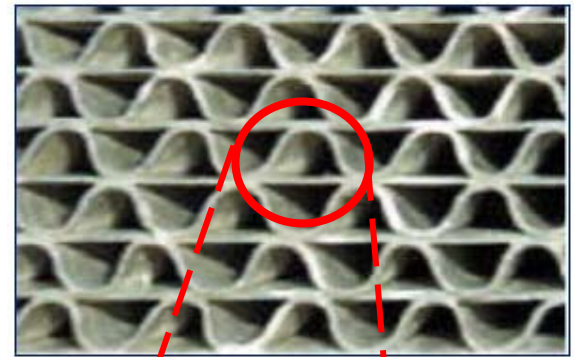
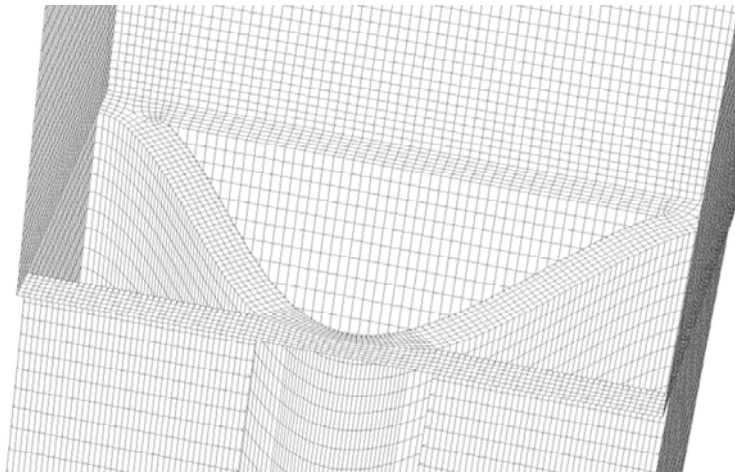
- For 24 and 48 hours average deposition rate more or less constant
  - Tendency of slight maximum after 15-25 cm
- After 78 and 120 hours average deposition rate increases in the first part of the monolith
  - Supports hypothesis that streamlining increases  $Re$  and raises deposition rate

# Quantitative discussion of dust deposition

- Overall mass deposition after 158 hours approx. 30.3 g
- Overall average deposition rate corresponds to 0.2 g/h
- Overall mass based deposition efficiency of approx. 30%
- Local deposition rate decreases in axial channel direction from inlet to outlet
- Increase in deposition rate in the first half of the channels with time indicating flow transition in this channel zone
- Deposition rate constant in the second half of the channels with time indicating unchanged flow pattern

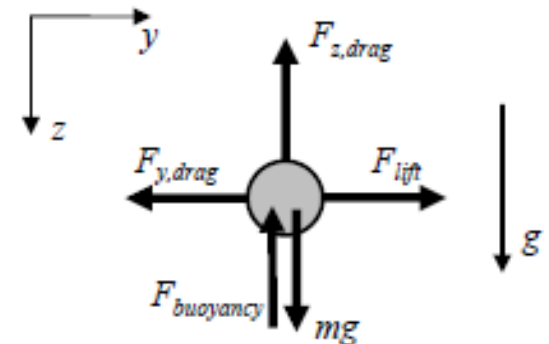
# CFD simulation of dust deposition

- Model grid with 517400 hexahedral cells

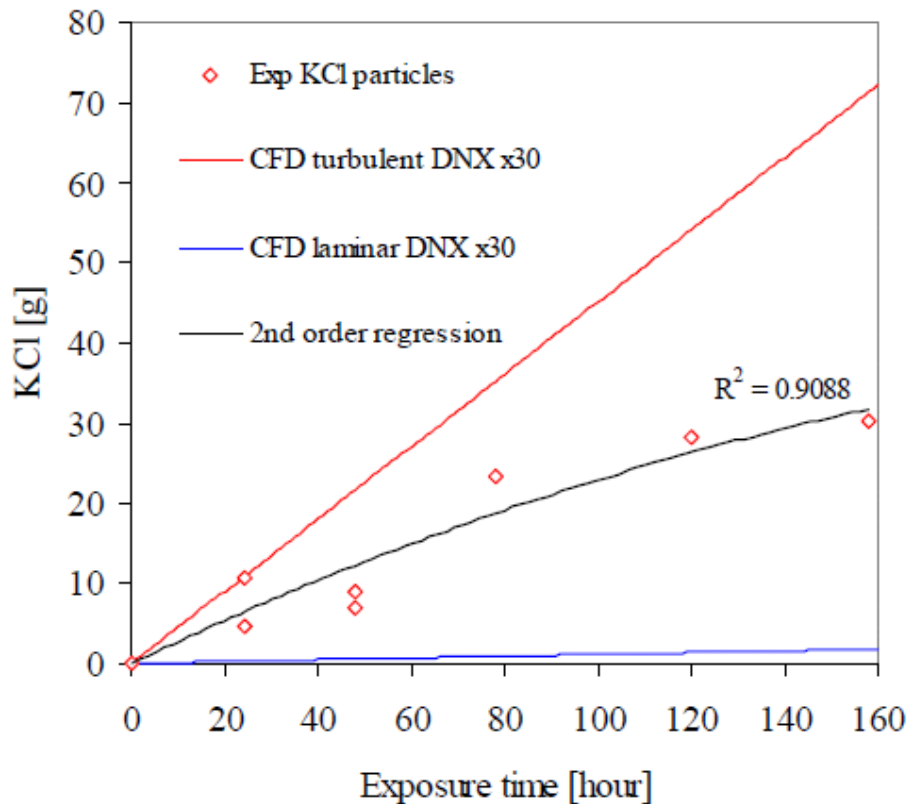


# CFD simulation model

- Steady-state transient laminar and turbulent CFD simulation for incompressible flow
- Continuity equation
- Reynolds-averaged Navier-Stokes equation
- $k$ - $\varepsilon$  turbulence model
- Discrete Particle Model (DPM) tracking 1.000.000 particles (corresponding to  $20 \text{ mg/Nm}^3$ ) with experimental particle size distribution
- Turbulent spreading of particles using the stochastic Random Walk Model (RWM)
- Electrostatic potential and electrostatic forces
- Balance of fluid dynamic forces
- Particle-Wall interaction



# Comparison between experiment and CFD



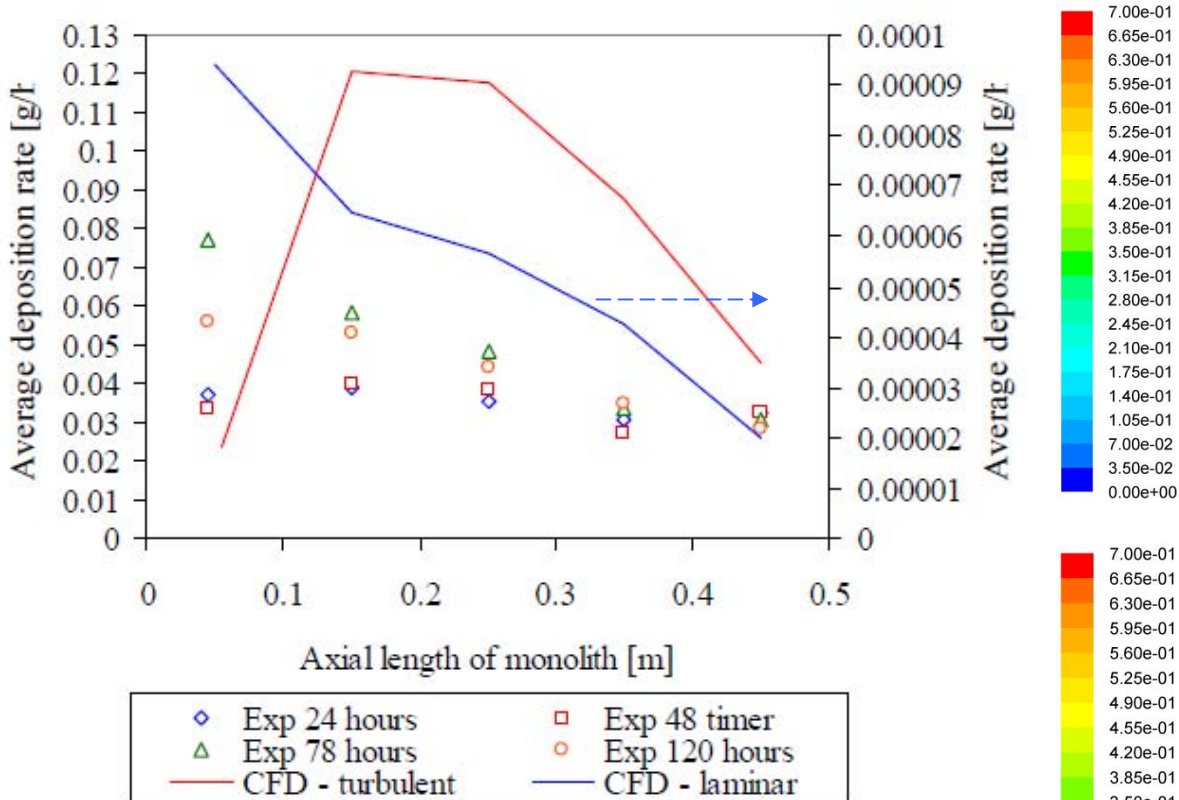
- Overall mass deposition on monolith over time
- Laminar flow underpredicts the deposition with a factor of approx. 17
- Turbulent flow overpredicts the deposition with a factor of approx. 2.4
- Confirms that flow must be in transition with turbulence being important in first part of monolith

# Deposition efficiency with CFD

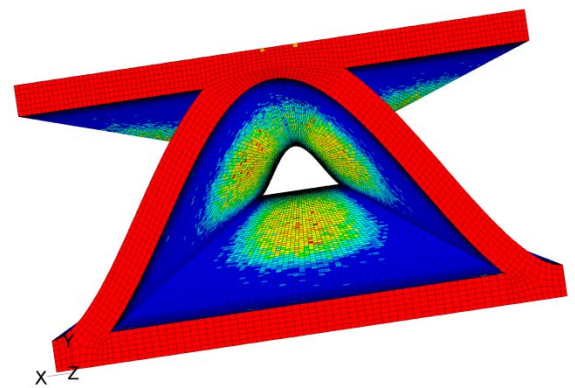
DNX x30 monolith	Laminar flow	Turbulent flow
	Deposition efficiency [%]	Deposition efficiency [%]
Top: Inertial impaction	1.32	2.27
Top: Brownian diffusion	0.01	-
Top: Inertial impact and turbulent diffusion	-	2.71
Top: Inertial impact, turbulent diffusion, Saffman lift and electrostatic forces	1.33	2.71
Channel: Inertial impaction, <u>turbulent diffusion</u> , Saffman lift and electrostatic forces	≈0	55.8
Top and Channel: Inertial impact, gravitation settling, turbulent diffusion, Saffman lift and electrostatic forces	1.33	59.8

- Dominating deposition mechanism in laminar flow is inertial impaction on the top
  - Responsible for 99% of deposition
  
- Dominating deposition mechanism in turbulent flow is turbulent diffusion in the channel
  - Responsible for 93 % of deposition
  
- Turbulent diffusion increases deposition on the top of the catalyst with 19%

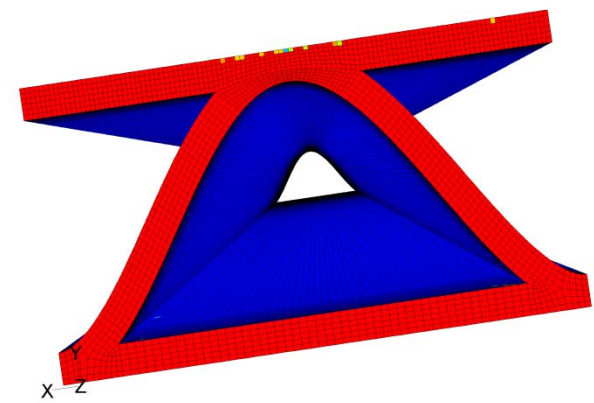
# Deposition rate in axial direction



Assuming turbulent flow



Assuming laminar flow



- Decreased particle conc. in channel inlet due to deposition at the entrance
- Fully developed flow in the 2nd half of the monolith

# Conclusions from PhD project

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- Deposition pattern ('volcano' type) created in the pilot setup experiments has also been observed at full-scale low-dust applications
- Deposition pattern observed for submicron particles
- Visual inspection show development of deposit at the top and in the monolith channels
- Average deposition rate highest at the top (after streamlining) and decreasing into the channel

# Conclusions from PhD project

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- Laminar flow underestimates particle deposition
- Turbulent flow overestimates particle deposition
- Flow must be in transition from turbulent to laminar through the monolith
- Flow transition occurs over half the monolith length, approx. 25 cm
- CFD with constant flow regime under-/overestimates the deposition compared to the experiment due to real flow transition

# Conclusions from PhD project

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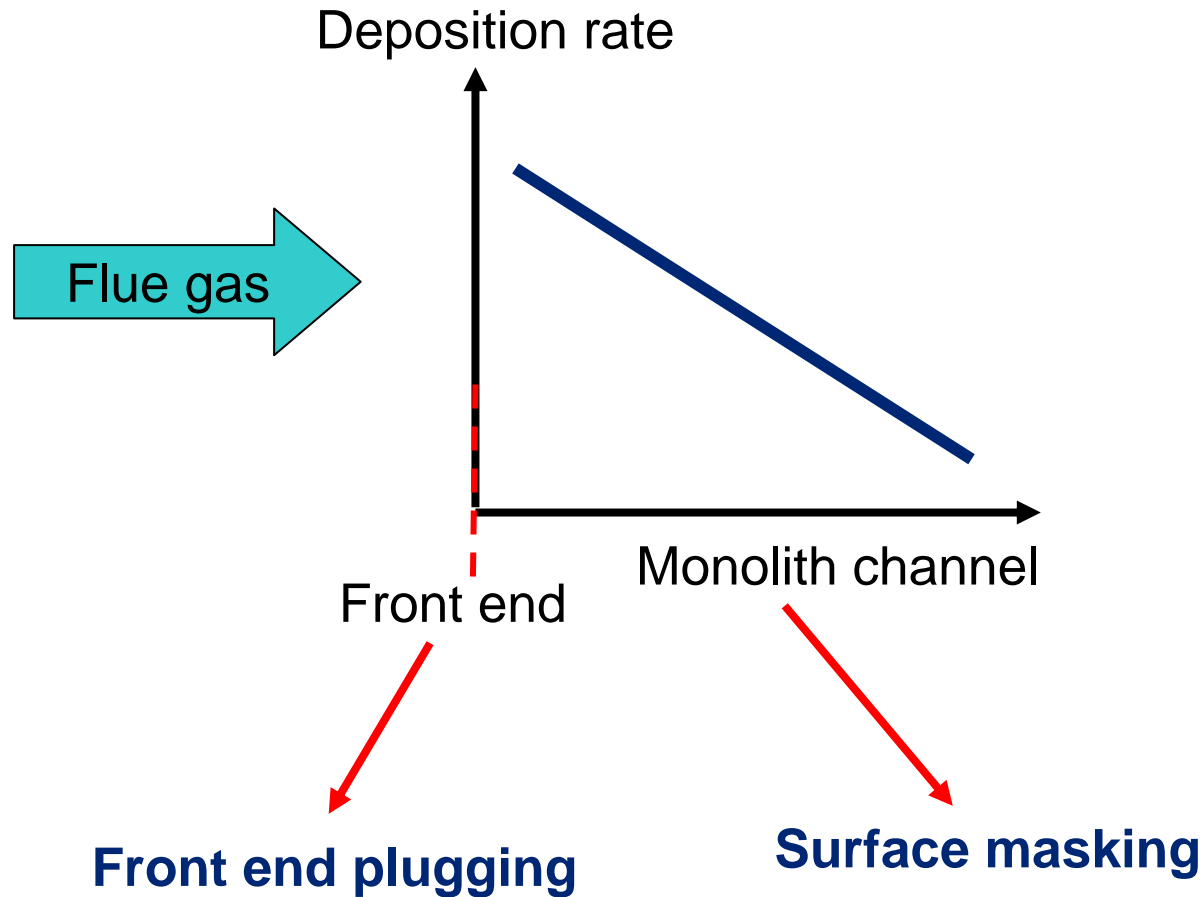
- Turbulence must be responsible for most of particle deposition
- Inertial impaction and turbulent diffusion responsible for deposition on top of the monolith
- Turbulent diffusion responsible for deposition in the monolith channels
- Turbulence upstream the monolith is an important factor enhancing particle deposition due to increased mass transport to the monolith wall
  - At least one order of magnitude higher compared to laminar flow upstream the monolith



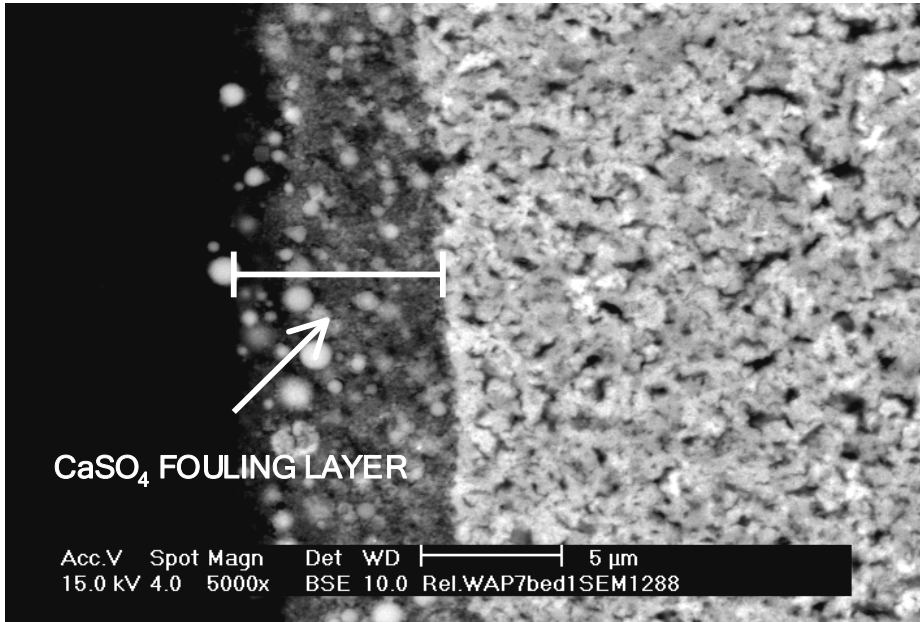
# Fly ash deposition in Low-dust SCR DeNO<sub>x</sub> Industrial experience

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# Fly ash deposition – practical challenges

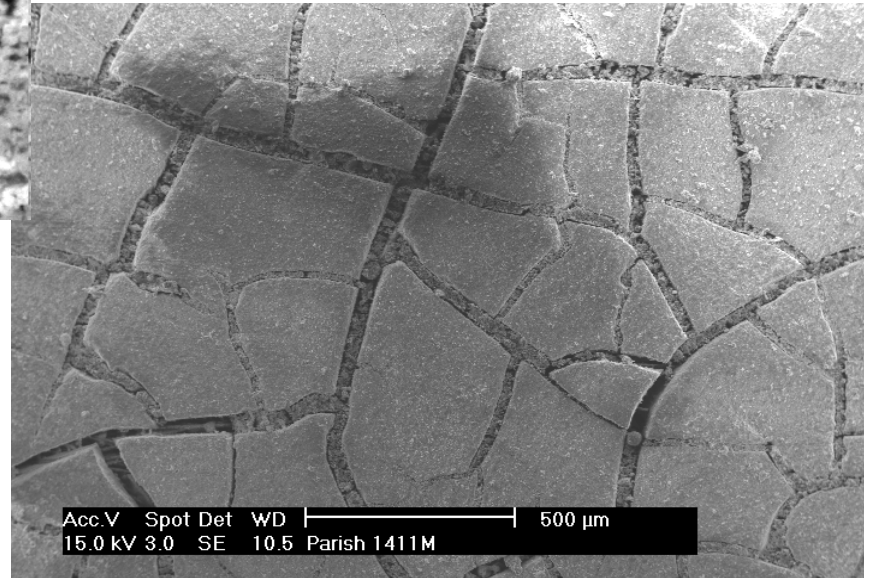


# Deposition of sub- $\mu$ particles in the channel

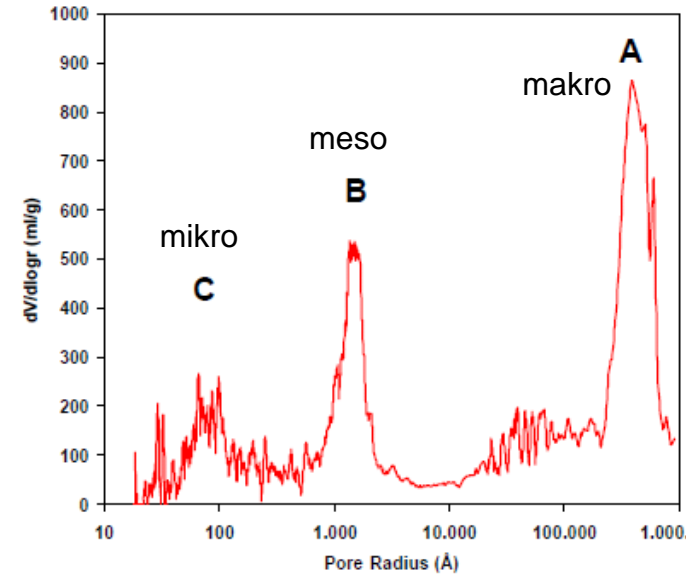
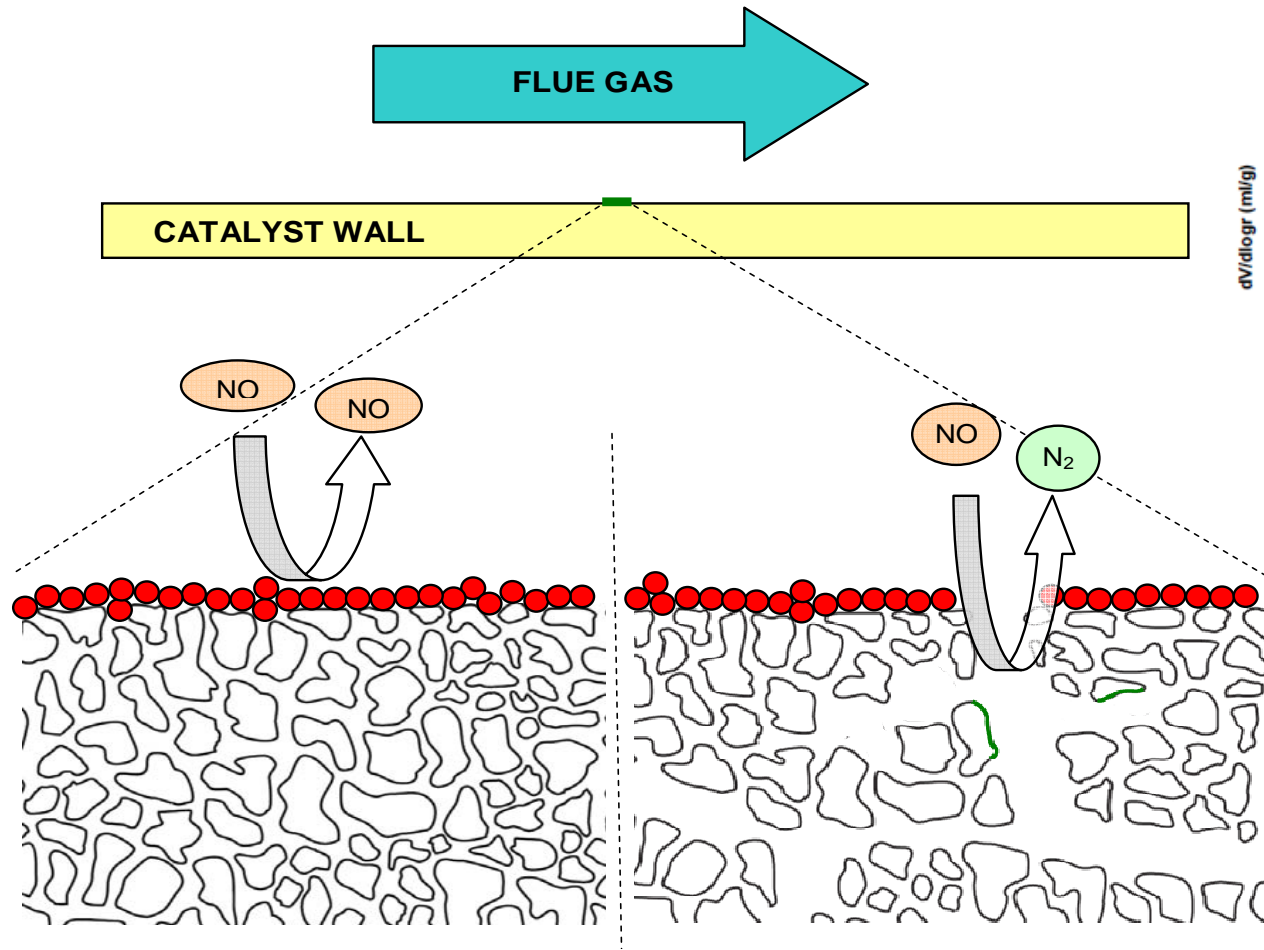


Surface fouling layer

Deposition of submicron particles



# Deactivation from surface masking



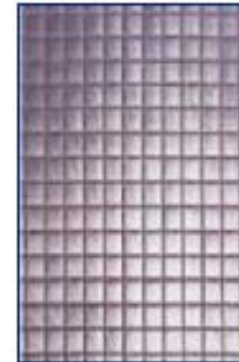
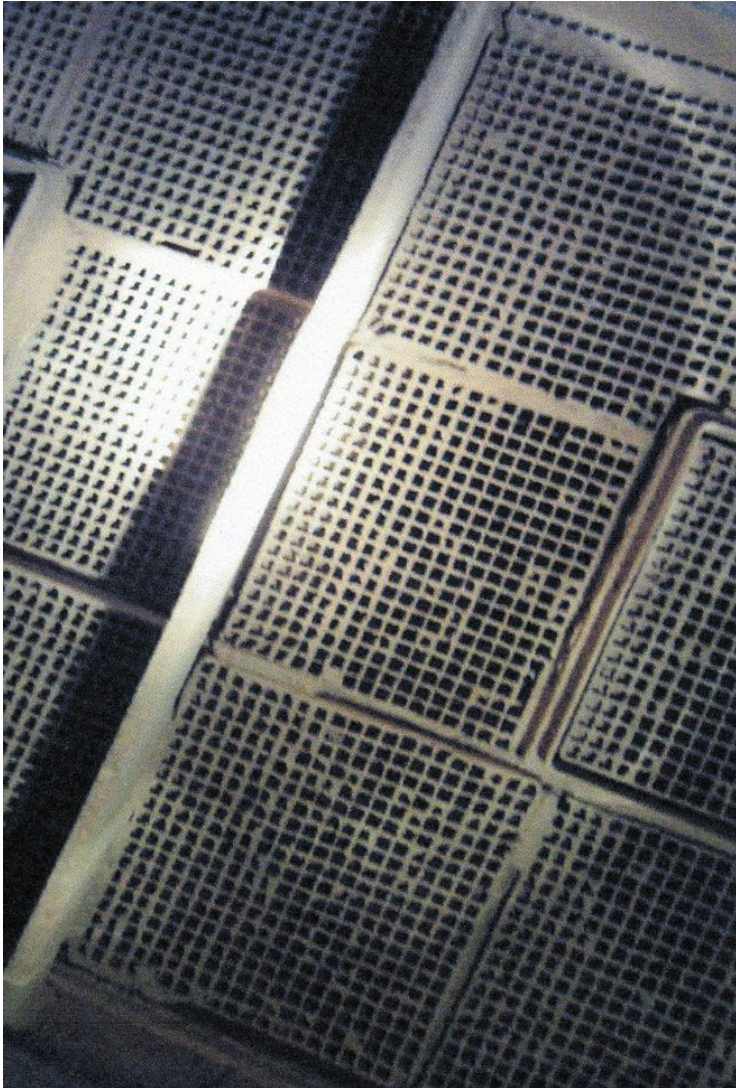
- Open pore structure with high capacity for dust uptake

# Proper pitch selection

- 16 MW grate-fired boiler burning wood
- Vertical flow SCR reactor after ESP
- Catalyst height 1.62 m
- Dust load specified for SCR design  $< 15 \text{ mg/Nm}^3$
- Catalyst pluggage at inlet side with chosen pitch within few month
- Actual dust load observed close to  $1000 \text{ mg/Nm}^3$
- Increased catalyst pitch required
- No increase in pressure drop since pitch change



# Fly ash deposition and pitch geometry



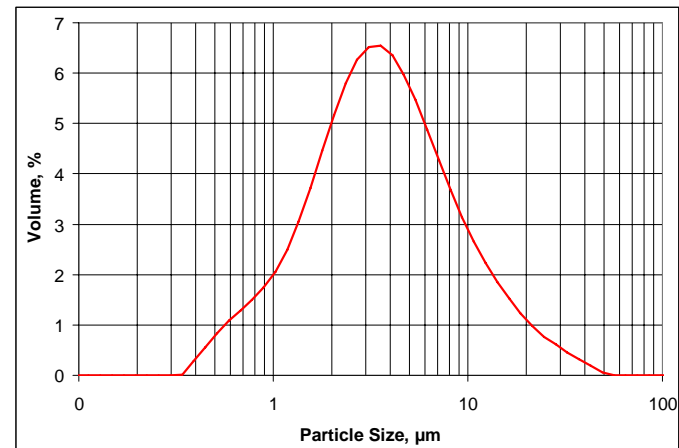
## Under operation with fly ash:

- Optimization of flow geometry to **cylindrical shape**, i.e. minimum surface-area-to-volume ratio for minimum friction
- **Minor importance of initial geometry**

# 'Volcano' build-up



- Low-Dust DeNOx in a refinery, DNX-939
- **No** soot blower or sonic horn
- Pressure increase/plugging after change in **ESP operation**
- 'Volcano' build-up in the top
- Plugging in the top
- Channels are free



# 'Volcano' build-up – ESP operation

- Some considerations
  - Change of ESP knocking frequency to continuous mode (due to transformer problems)
  - Decrease in dust density from ESP (from 300 kg/m<sup>3</sup> to 170 kg/m<sup>3</sup>)
  - Potential impact on dust deposition on ESP
    - Usual non-continuous mode: rough surface due to dust build-up → enhanced turbulence → enhanced mass transport and deposition
    - Continuous mode: less pronounced
  - *Remember trouble with new heat exchanger in experimental section (analogy)*
- Consequently, for DeNOx design
  - Increased dust load beyond design specifications

# Particle charging from ESP

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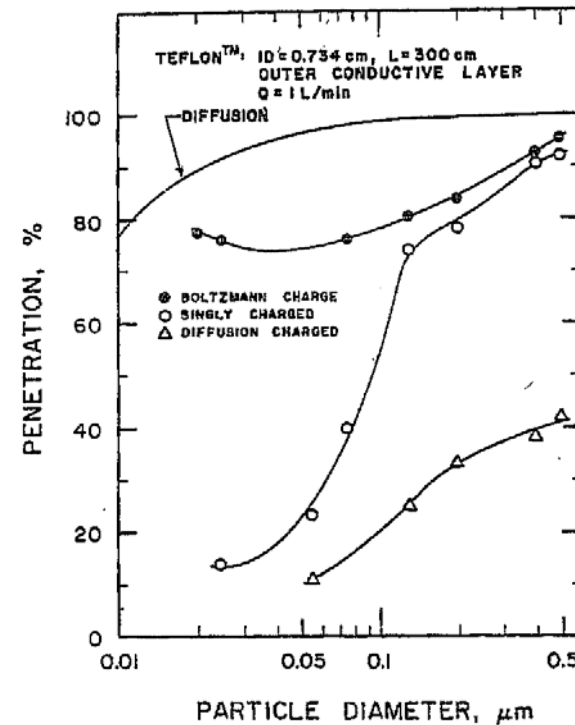
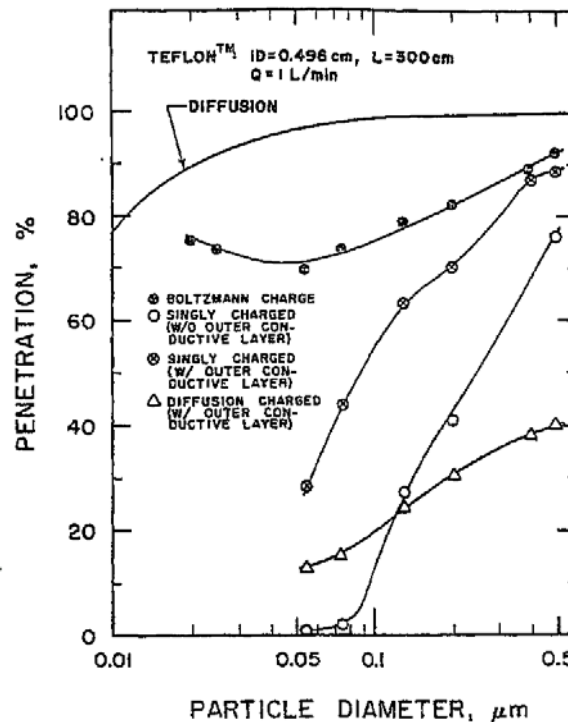
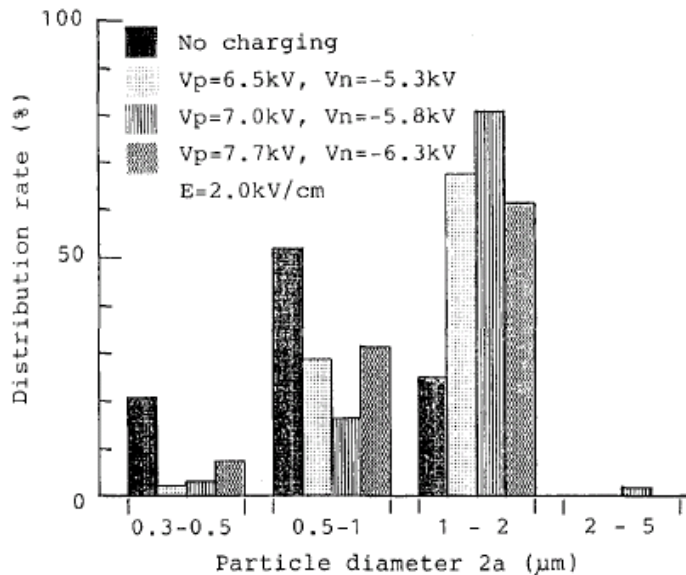
- Electrostatic charging of dust particles from ESP observed
  - where residence time  $<$  charge relaxation time
- Electrostatic charging enhances particle deposition and agglomeration to larger chunks (in analogy with ESP)

# Particle charging from ESP

- Electrostatic charging has pronounced effect on submicron particles

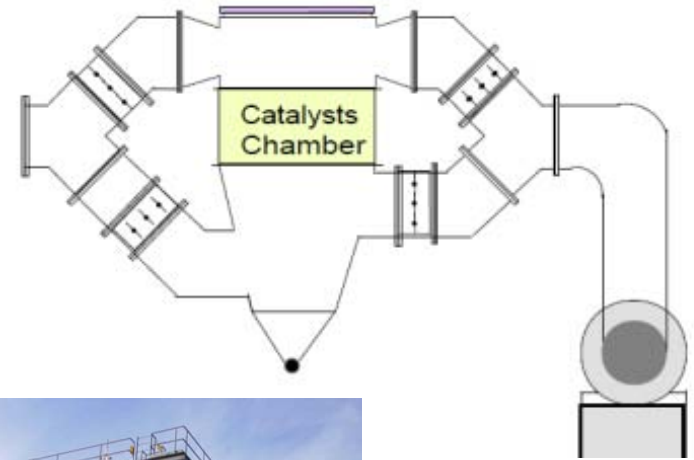
## Enhanced deposition of submicron particles

### Enhanced agglomeration



# Catalyst cleaning – reverse flow

- 16 - 22 MW grate-fired boilers
- Fuel: wood chips
- SCR installed in 2008
- Low-dust after ESP
- Vertical or horizontal flow
- No soot blowers
- Catalyst cleaning
  - reversing flow 1-2 times per diem



# Catalyst cleaning – reverse flow

## ■ Some considerations

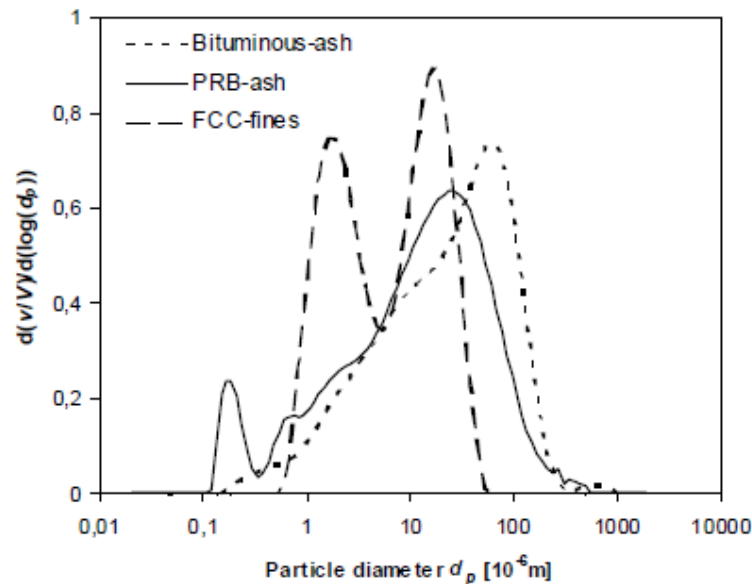
- Particle detachment when applied forces overcome adhesion forces (van der Waals forces, electrostatic forces)
- For small particles  $< 10 \mu\text{m}$ , adhesive force much greater than removal forces (gravitational, vibrational, centrifugal)
  - Adhesion  $\sim D_p$  and removal forces  $\sim D_p^2, D_p^3$
- However, layer of particles less than  $10 \mu\text{m}$  easily dislodged in large  $0.1 - 10 \text{ mm}$  chunks

## ■ Consequently, for reverse flow cleaning

- Cleaning of channel pluggage and removal of large dust agglomerates possible
- Submicron particle detachment from channel wall not possible

# SCR DeNO<sub>x</sub> in Fluid Catalytic Cracking

- Low-dust compared to coal (10-100 kg/h)
- ESP can be avoided with Haldor Topsøe design
- FCC catalyst fines in the flue gas below 10 μm, and large fraction of fine particles around 1 μm



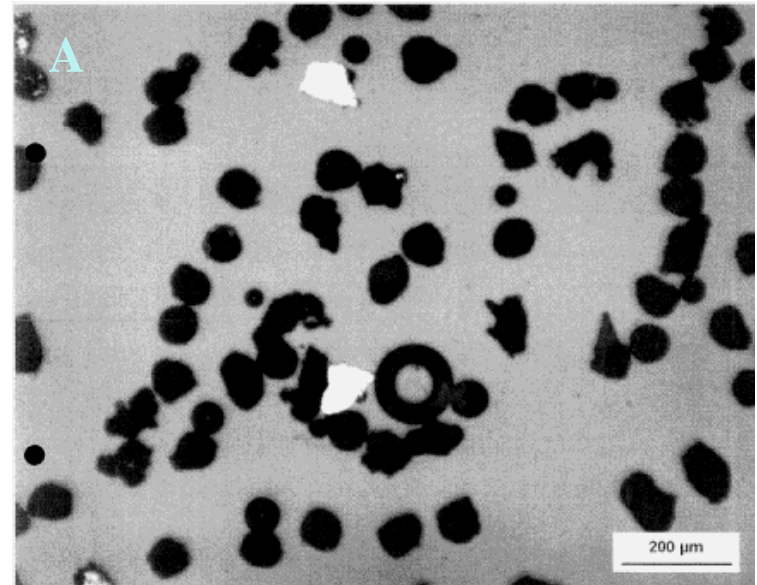
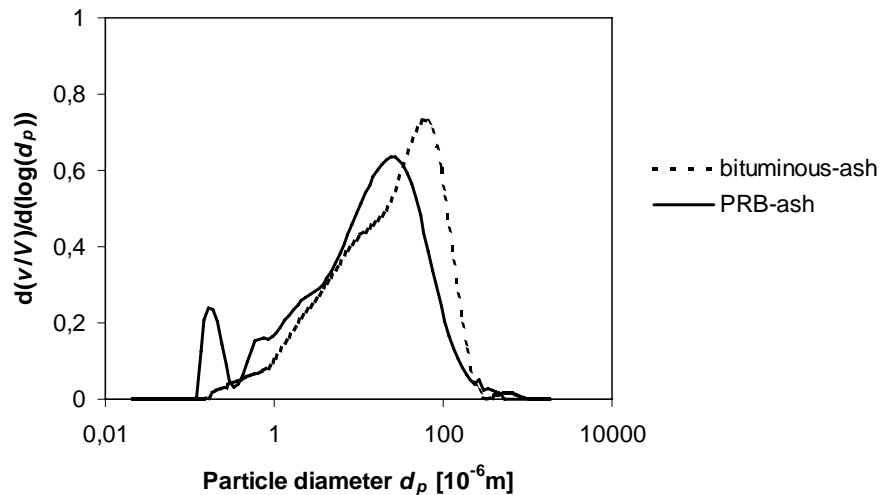
# SCR DeNOx in Fluid Catalytic Cracking

- Sticky nature of the dust requires a wide-pitch catalyst
- Dust build-up on the top grid wire mesh and on the catalyst front edge
- Cleaning with rake-type soot blowers with superheated steam or compressed air (1-3 times per day)
  - Sonic horns not able to remove the dust build-ups



# Selv-cleaning effect of large particles

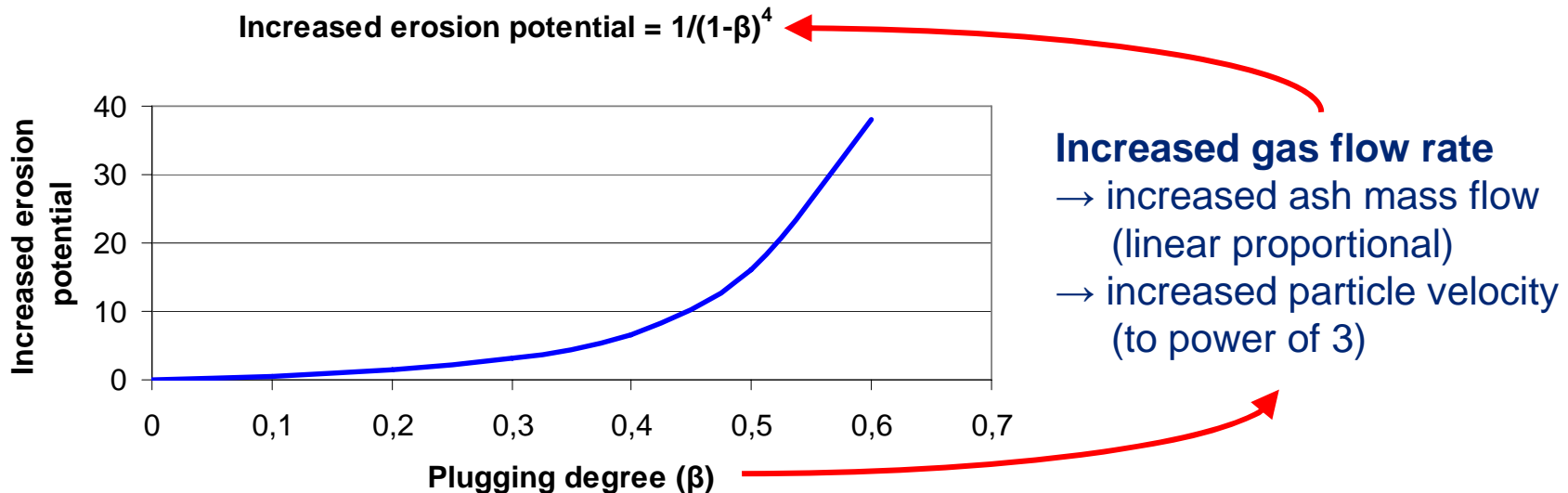
- Larger fly ash particle with high inertia ( $> 20 \mu\text{m}$ ) have a cleaning effect by detaching and re-suspending deposited particles



- New challenge with **large particle ash**:  
Clogging and Erosion  $\rightarrow$  High-dust applications before ESP

# Erosion of SCR DeNOx catalyst

- Clogging the channels by too large particles enhances erosion for all types of DeNOx catalysts



- Impacting not only front edge but whole monolith length
- Careful selection of pitch and wall thickness, as well as correct design of DeNOx plant of paramount importance



# Fly ash deposition in SCR DeNOx Summary and conclusion

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# Summary

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- Volcano-type deposition and streamlining in the inlet
- Turbulence enhances particle deposition
- Shaping of channel geometry to cylindrical shape
- Pluggage due to fly ash lumps on the inlet side
  - Proper soot blowing helps keeping the front end clean
- Fly ash fouling on the catalyst surface in the channels
  - Impeding gas diffusion and reaction
  - Open pore structure with high dust capacity limits pore diffusion restrictions

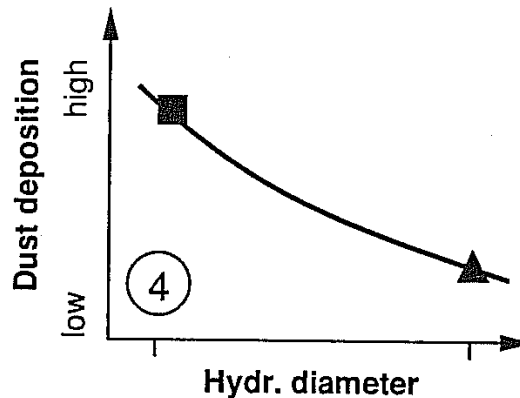
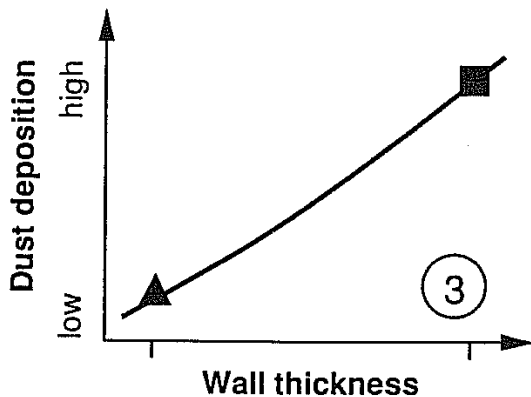
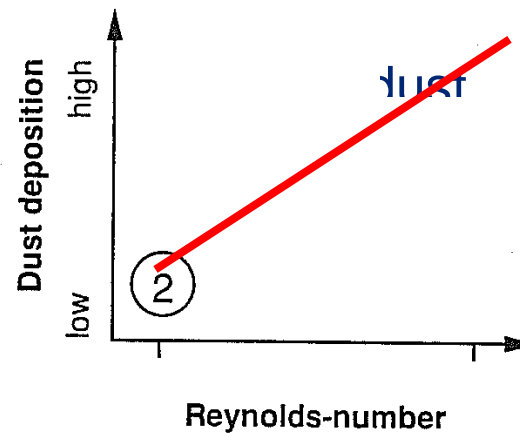
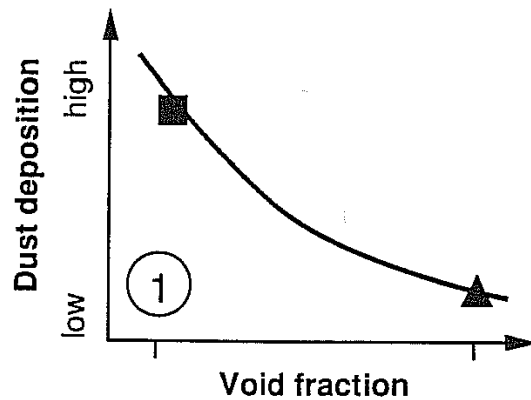
# Summary

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- Careful choice of pitch size
- Electrostatic charging (from ESP) affects submicron particles (enhanced agglomeration and deposition)
- Reverse flow cleaning can remove fly ash lumps
- Self-cleaning effect of larger particles
  - However, issue of clogging and erosion with LPA
- CFD a powerful tool for qualitative trends and to identify critical areas of dust deposition

# Dust deposition dependence for Low-dust

- as function of geometric and fluid dynamic parameters



# Pitch selection versus dust load and activity

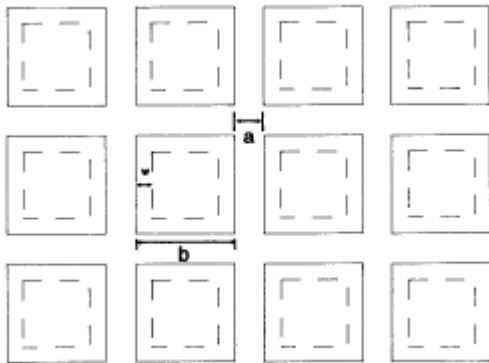


Figure 1. Monolith channel geometry.

## Growth of fouling layer

$$dw / dt = k_{m,f} C_f / \rho_f$$

$k_{m,f}$  = mass transfer coefficient

Topsøe DNX® catalysts

Catalyst type	Channel size, mm	Typical applications
DNX-HD	6.5 - 9.0	High-dust coal-fired boilers
DNX-MD	4.7 - 5.6	Medium dust coal-fired boilers Diesel engines
DNX-LD	3.4 - 5.1	Low dust oil and gas-fired boilers Waste incinerators
DNX-ND	2.7	Dust-free flue gases from combustion of natural gas

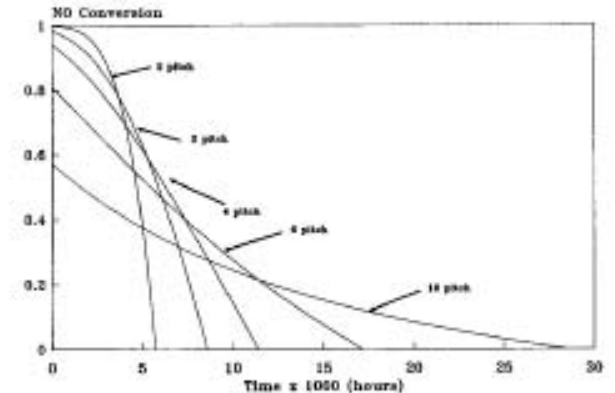


Figure 2. NO conversion versus time for varying catalyst pitch.

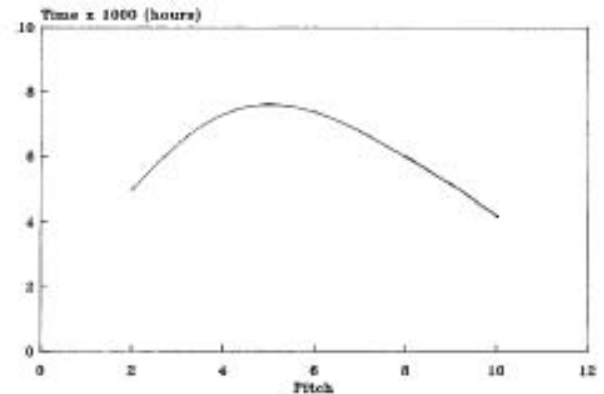


Figure 3. Time to reach NO conversion of 40% for varying catalyst pitch.

# Thank you very much for your attention!

