Fundamental Modeling and Experimental Studies of Acicular Mullite Diesel Particulate Filters

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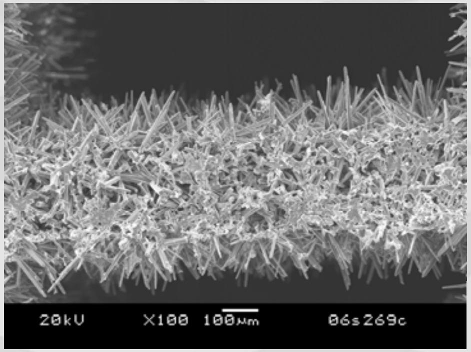
Acicular Mullite (ACM) DPF substrate



- Unique ceramic filter material
- Under continuing development at Dow Automotive
- Introduced at SAE 2004 World Congress (SAE 2004-01-0955)
- Employed by Audi R10 TDI
  - 24 Hours of Le Mans winner 2006, 2007, 2008
  - 12 Hours of Sebring winner 2006, 2007
  - 5.5 L diesel engine, 650hp

Dow Automotive / PNNL CRADA

- Cooperative Research and Development Agreement between Dow Automotive and PNNL began in summer of 2005
- PNNL goal has been to assist in understanding parameters affecting filter performance through:
  - Fundamental experiments at EESL
  - Pore-scale modeling using PNNL supercomputers
- Communication maintained via on-site visits, monthly teleconferences, and Sharepoint website

Exhaust Emissions Science Laboratory

mpp2 supercomputer

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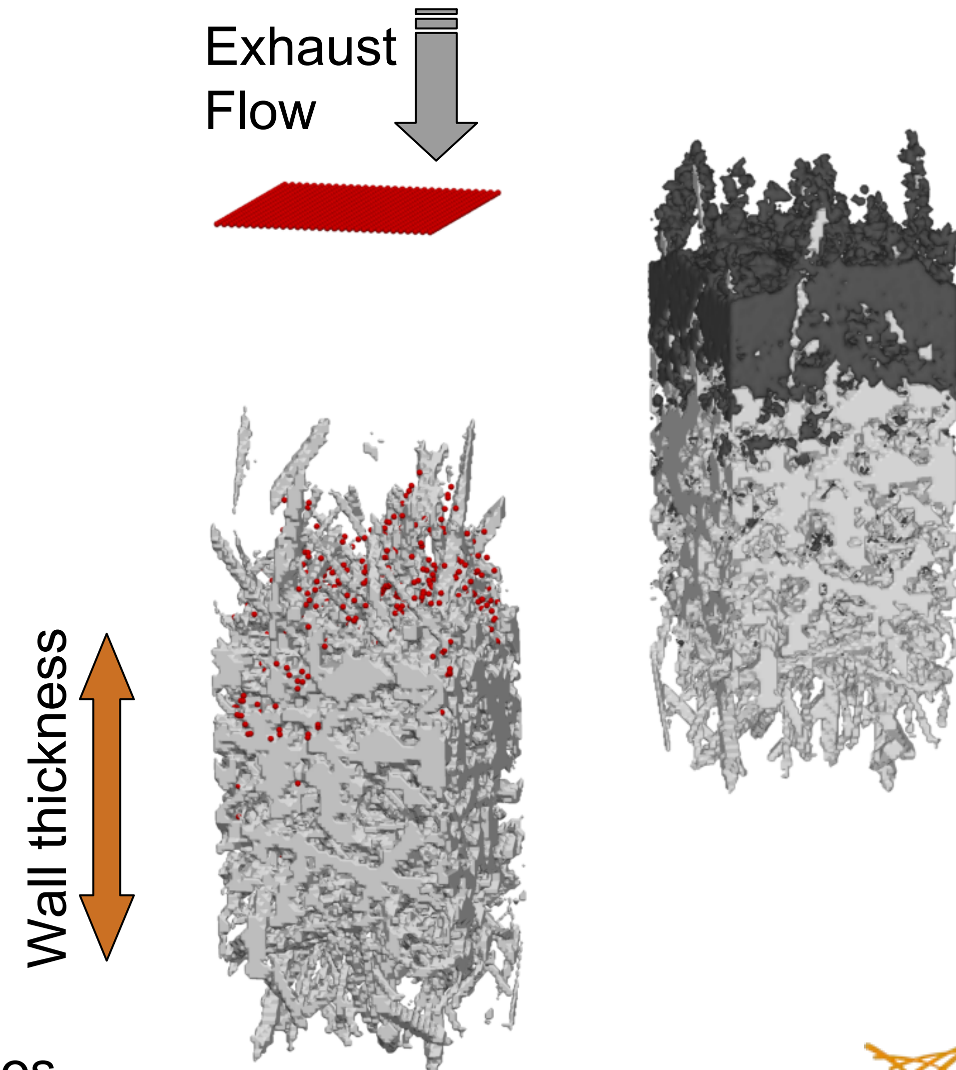
Pore scale filtration simulations

- 3D digital microstructure reconstructed using statistical information from micrographs
- Exhaust velocity and pressure fields continuously solved using the lattice-Boltzmann method
- Flight and deposition of individual particles simulated via drag forces and Brownian motion
- Goal is to provide fundamental insight, not directly model devices

Similar study: Konstandopoulos et al. SAE-2007-01-1131



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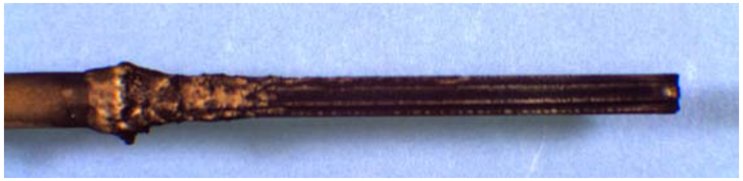
Fundamental DPF experiments

- Single-channel fluid dynamics and filtration
- Single-wall fluid dynamics and filtration
- Filtration tests with LD diesel soot (VW TDI)
- Filtration tests with surrogate particles
  - Aerosolized salts
  - Laboratory soot generator
- Bench reactor tests for soot and NO_X oxidation kinetics

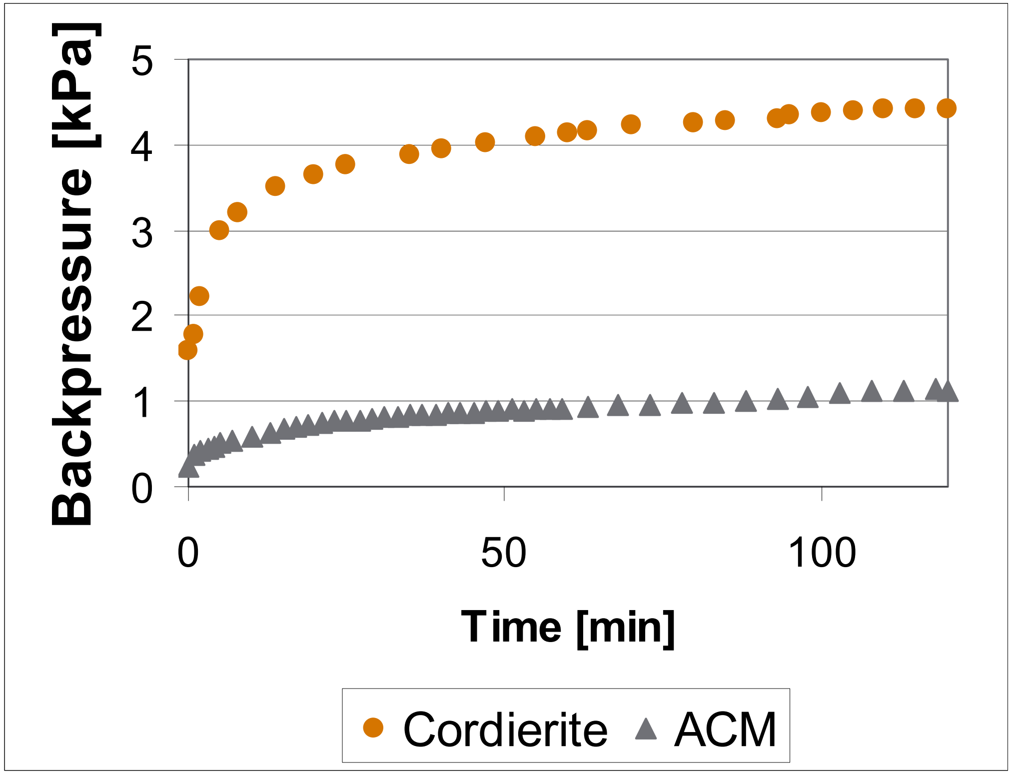


Loaded single-channel filter sample

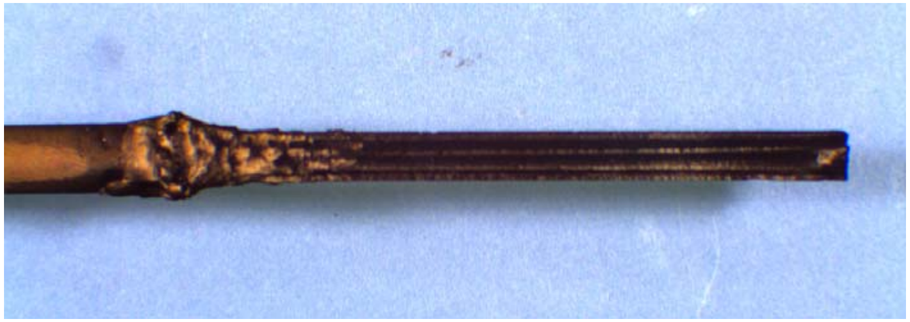
Single-wall filter sample

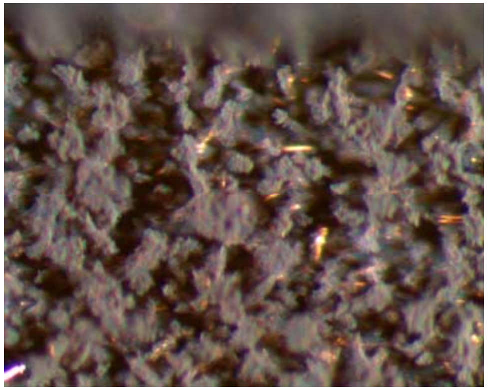
Bench reactor and furnace

Typical single channel soot loading data



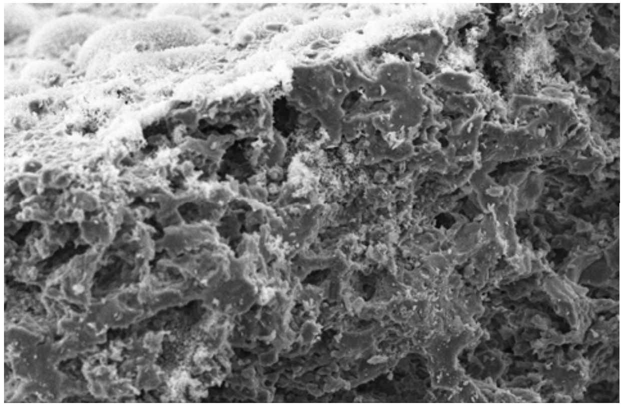
- Soot generated by VW TDI on chassis dyno
- Exhaust flow nearly normal to exterior channel surfaces
- ACM has lower clean backpressure
- ACM exhibits a less dramatic increase in backpressure before transition to cake filtration



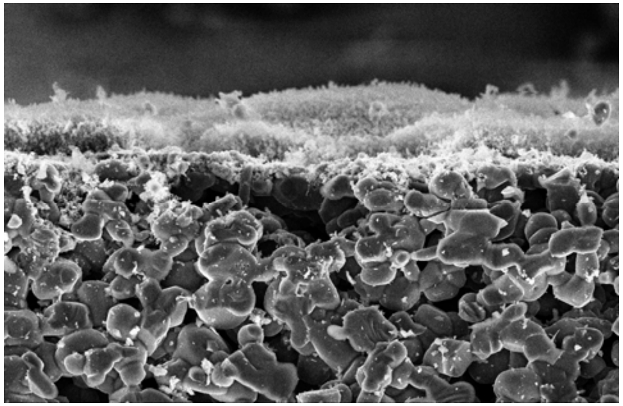


Filtration studies with aerosolized ammonium sulfate particles

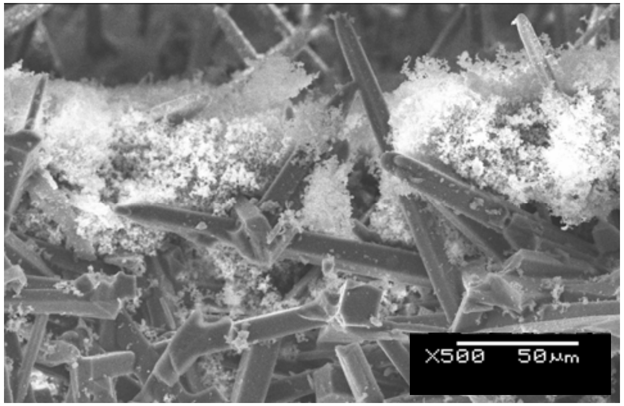
Cordierite

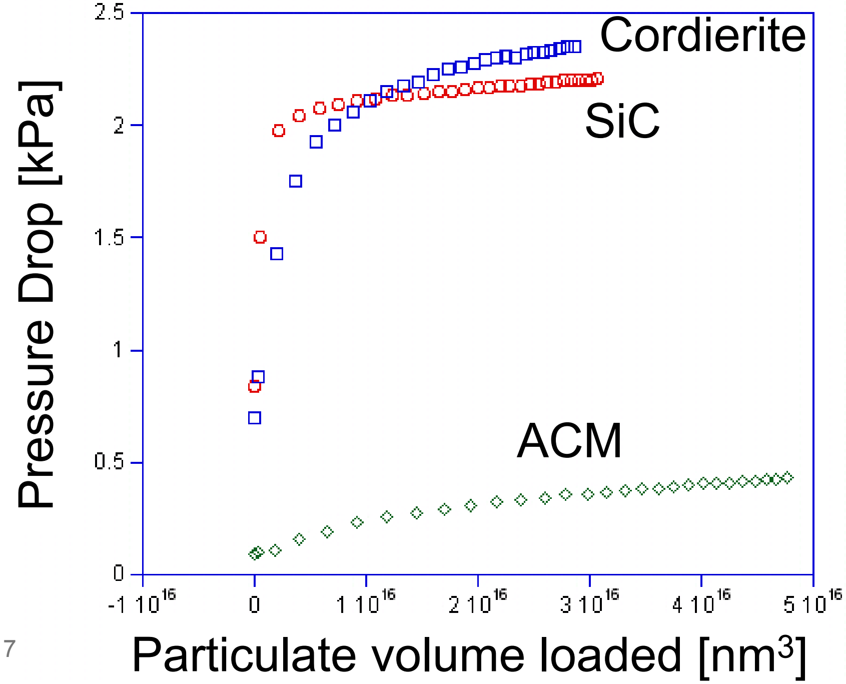


Silicon Carbide



ACM





- Roughly spherical particles are more consistent and less complex than real soot
- Particle size distribution can be altered
- Filters samples can be washed and re-loaded

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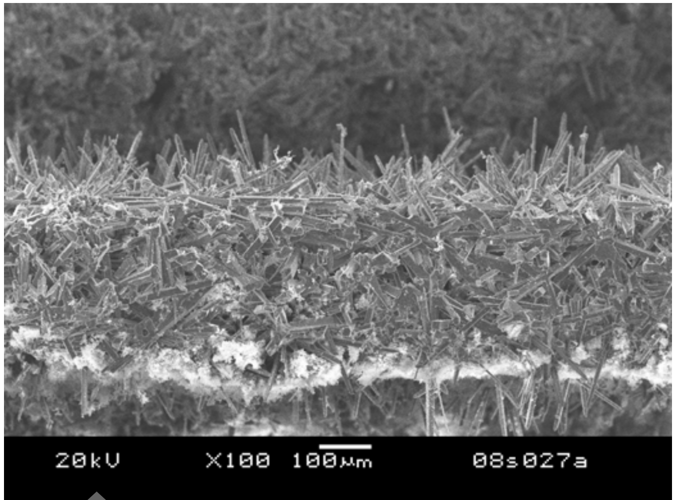
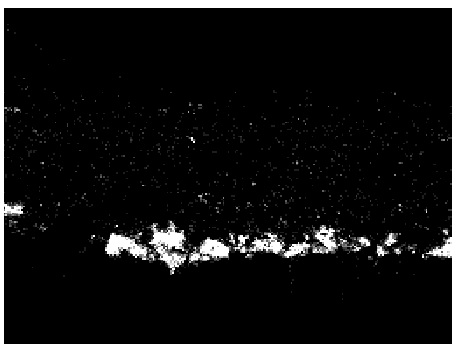
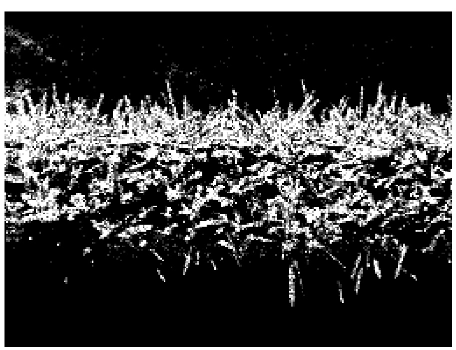
Wall penetration data from salt particle filtration studies



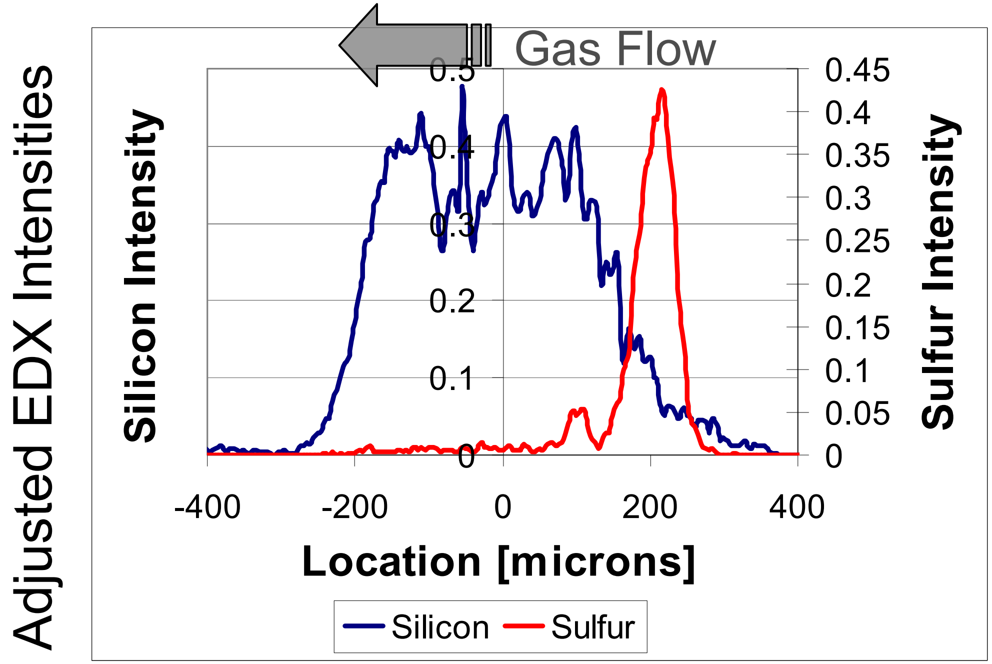
Gas Flow

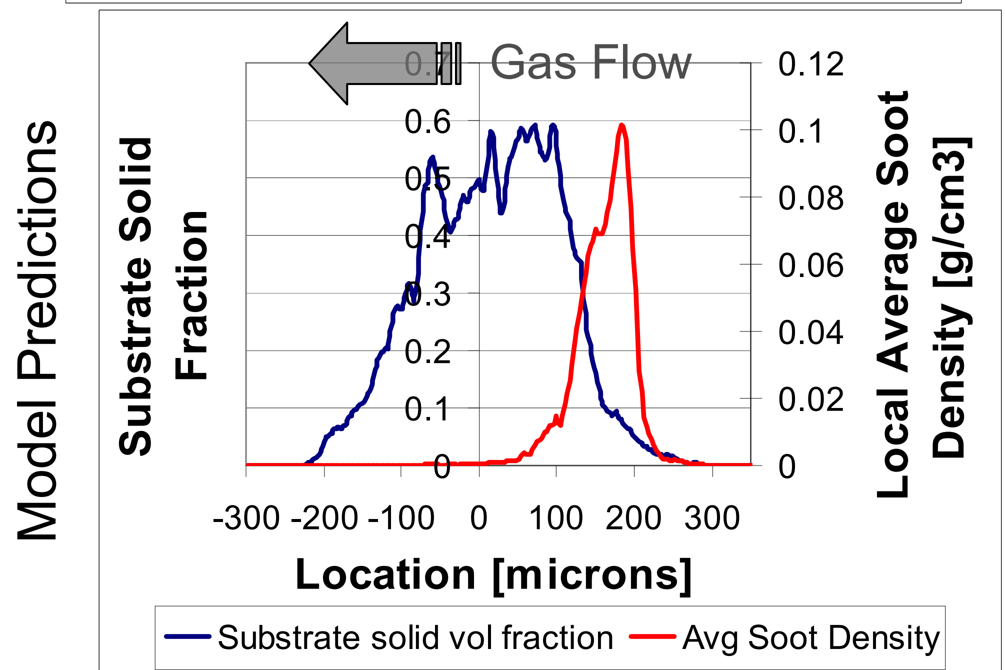


Sulfur EDX



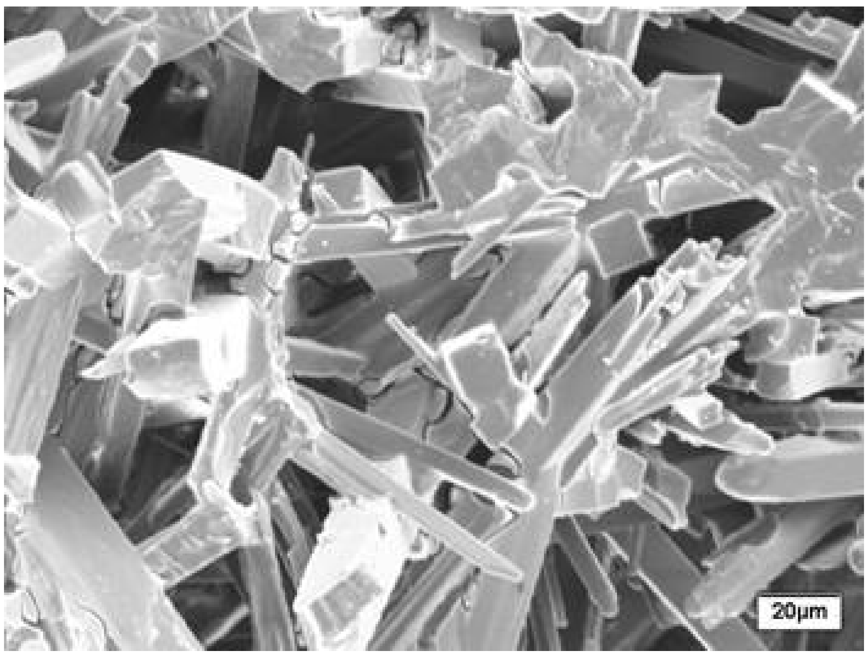
Silicon EDX



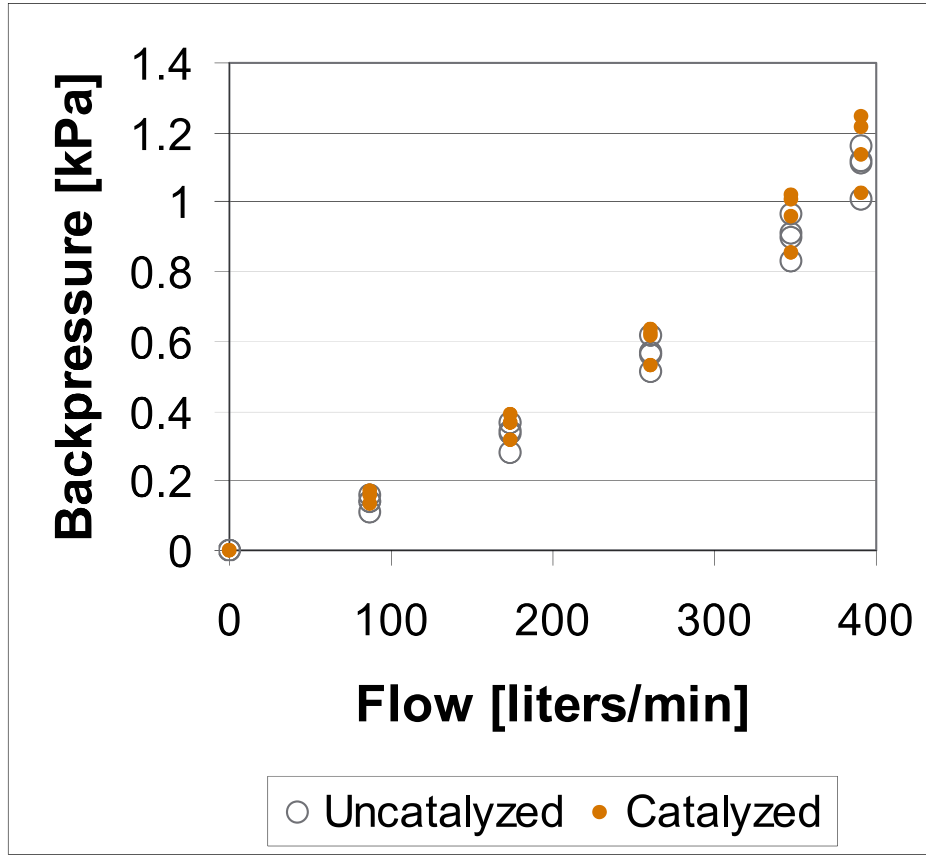


NO oxidation on catalyzed ACM

- Thin catalyst coatings have a very minor impact on clean back-pressure
- NO oxidation experiments indicated a turnover rate of ~7.0E-3 mol/mol Pt at 300 ppm NO and 300°C



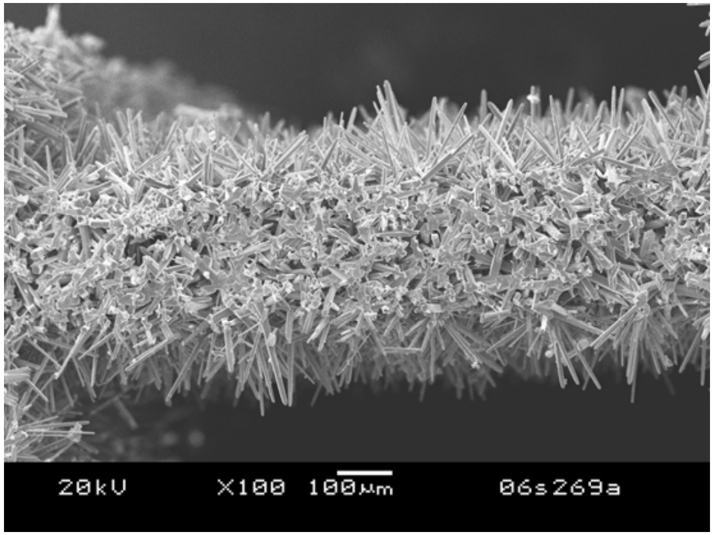
Catalyzed ACM

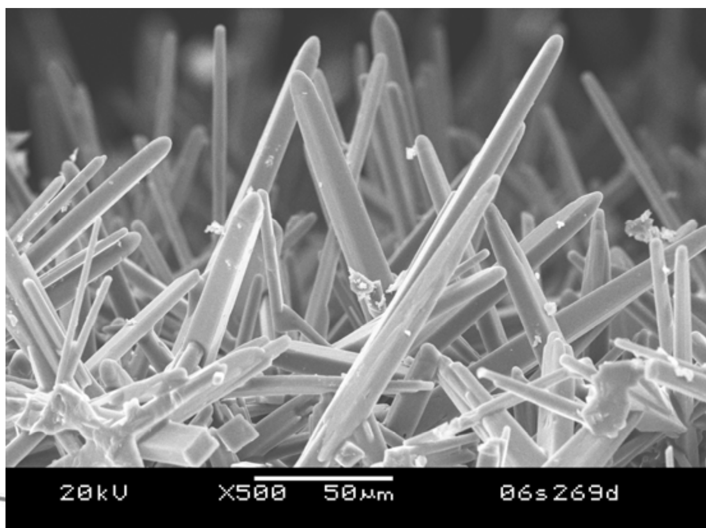


- A TOR of ~2.2E-3 mol/mol Pt was measured under similar conditions for Pt on gamma alumina by Mulla et al.

Journal of Catalysis 241 (2006) 389-399

Role of ACM micro-structure





- The most striking physical feature of the ACM substrate is the presence of mullite 'whiskers' or 'needles' protruding from the filter wall surfaces
- What role do the protruding structures play in determining filter performance?
- Filter simulations in ACM with the protruding needles removed were compared to simulations with the original geometry

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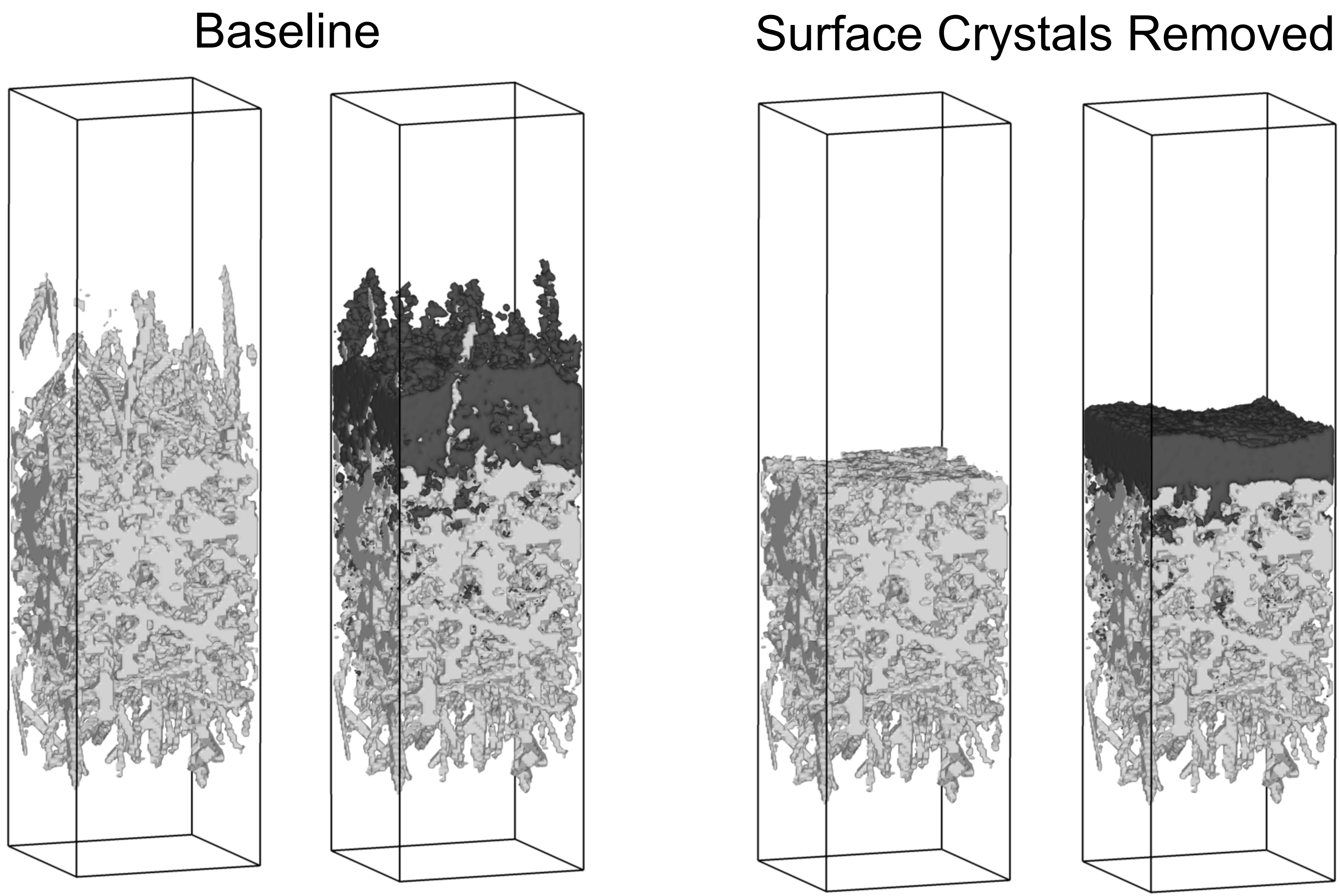
Simulated soot loading to approximately 5.25 g/L



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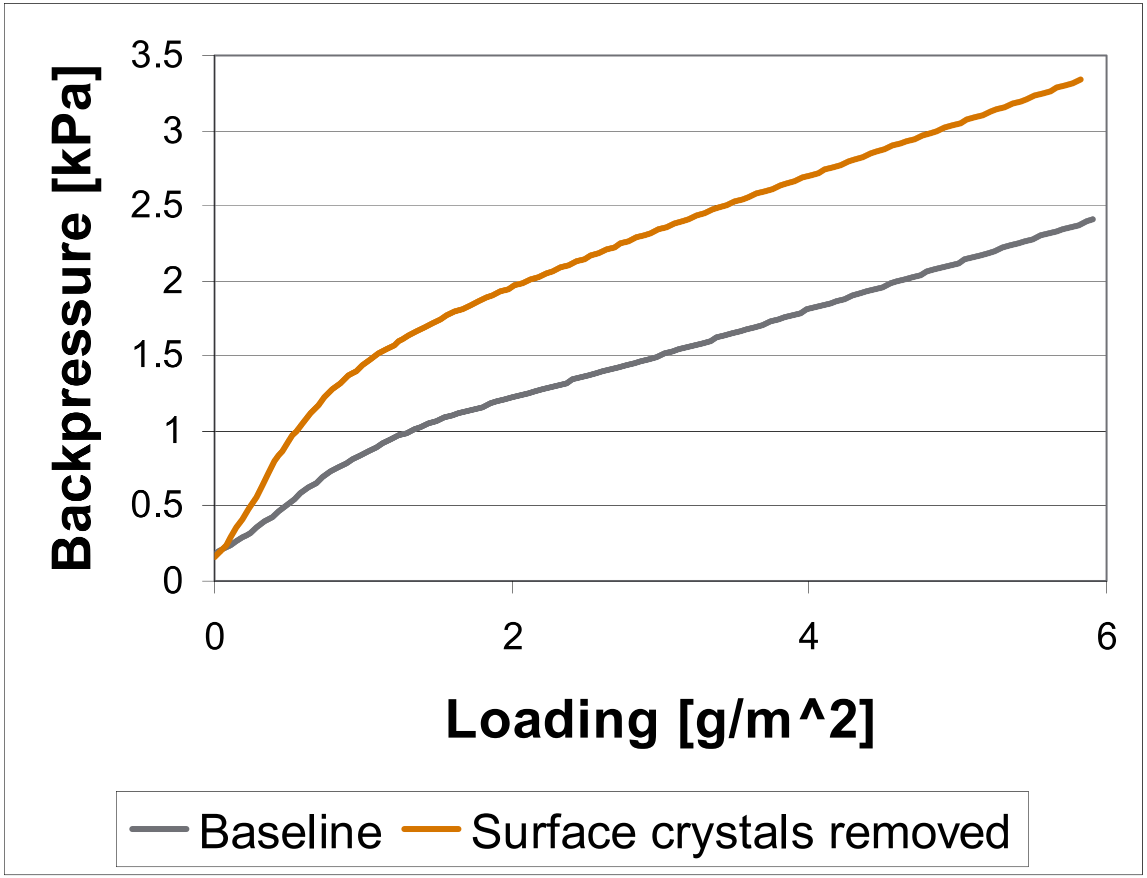
Back-pressures predicted by simulations



- Protruding needles have little effect on clean pressure drop for flow normal to the filter wall
- Δp slopes during cake filtration are slightly greater without needles
- Needles have the biggest impact during the transition to cake filtration

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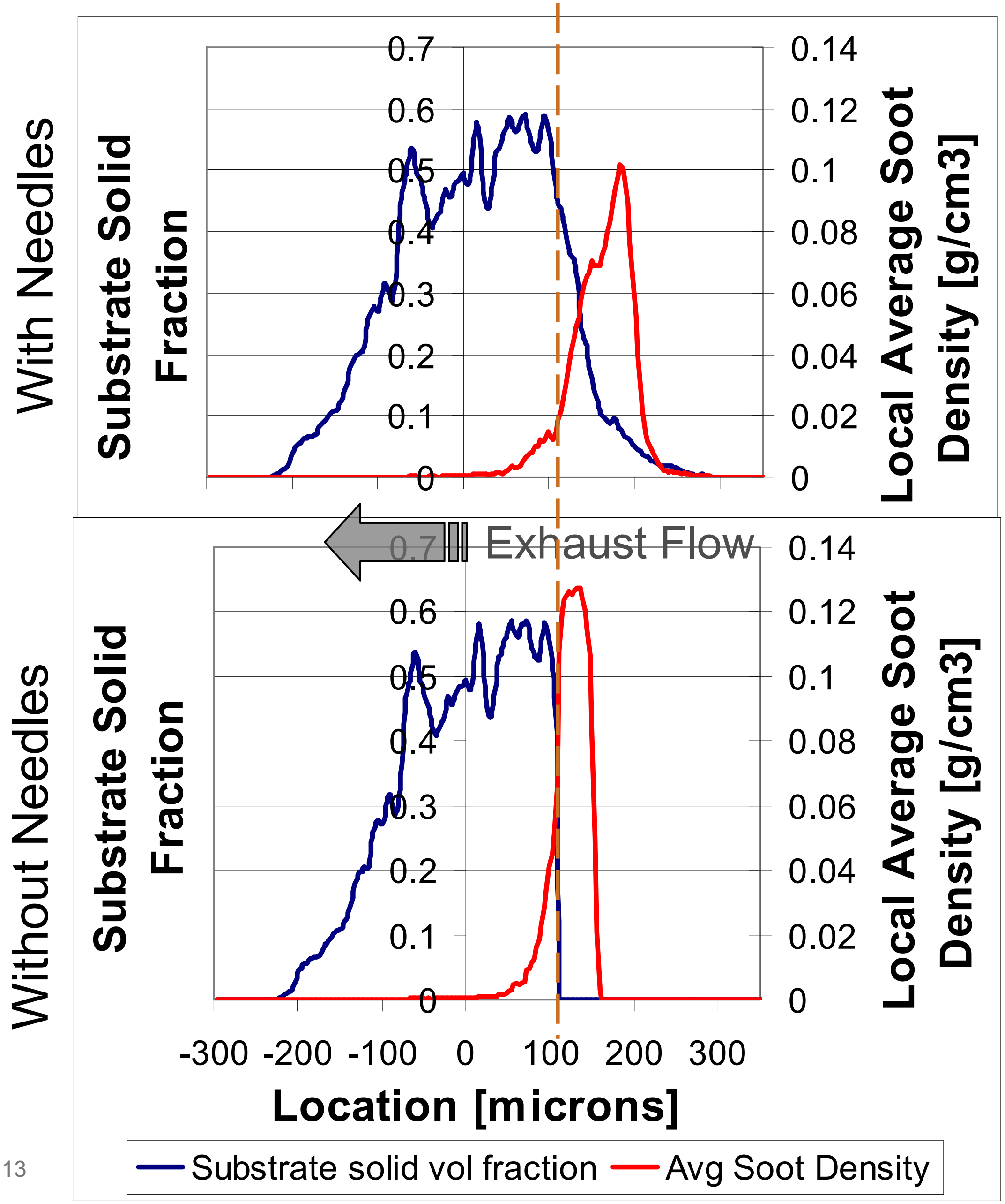
Predicted soot penetration into wall



- Needles result in a soot 'cake' with lower average density
- Removal of needles allowed more penetration of soot into the lower porosity core region of the filter wall
- Model does not include compression of soot cake under transient loading or flow conditions - could be an additional factor

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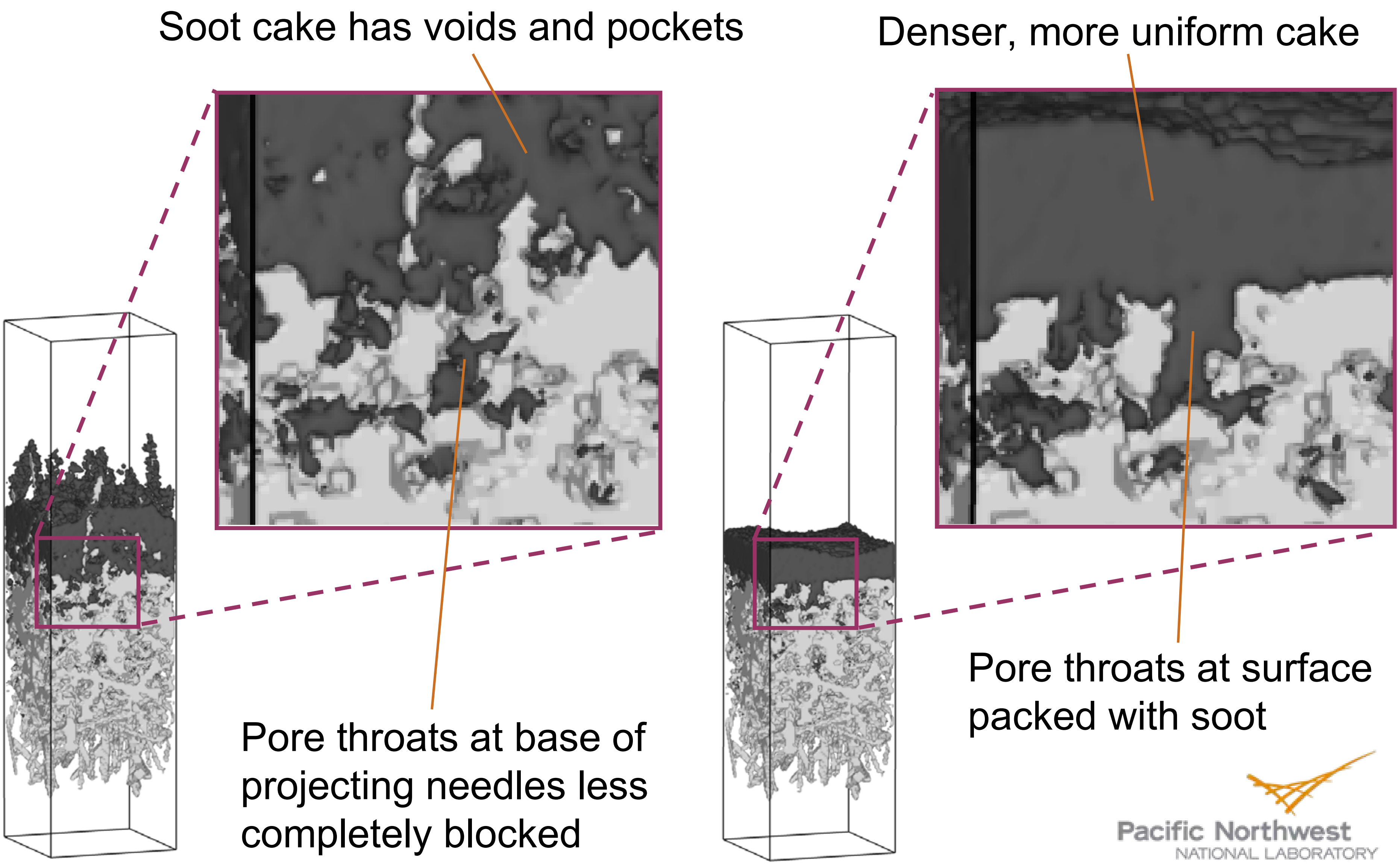
Close-ups of simulated soot deposits with and without projecting needles



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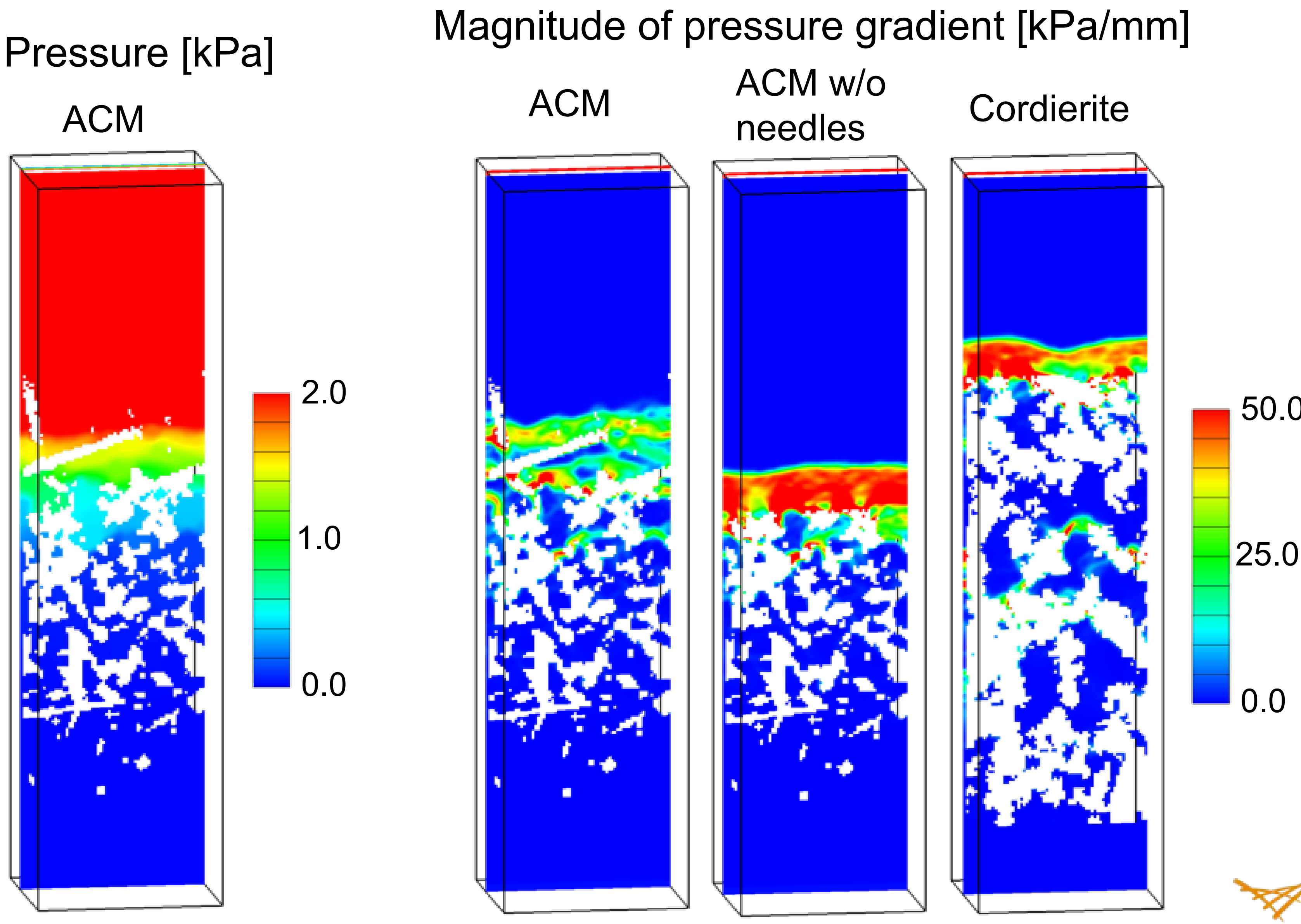
Pressure gradients



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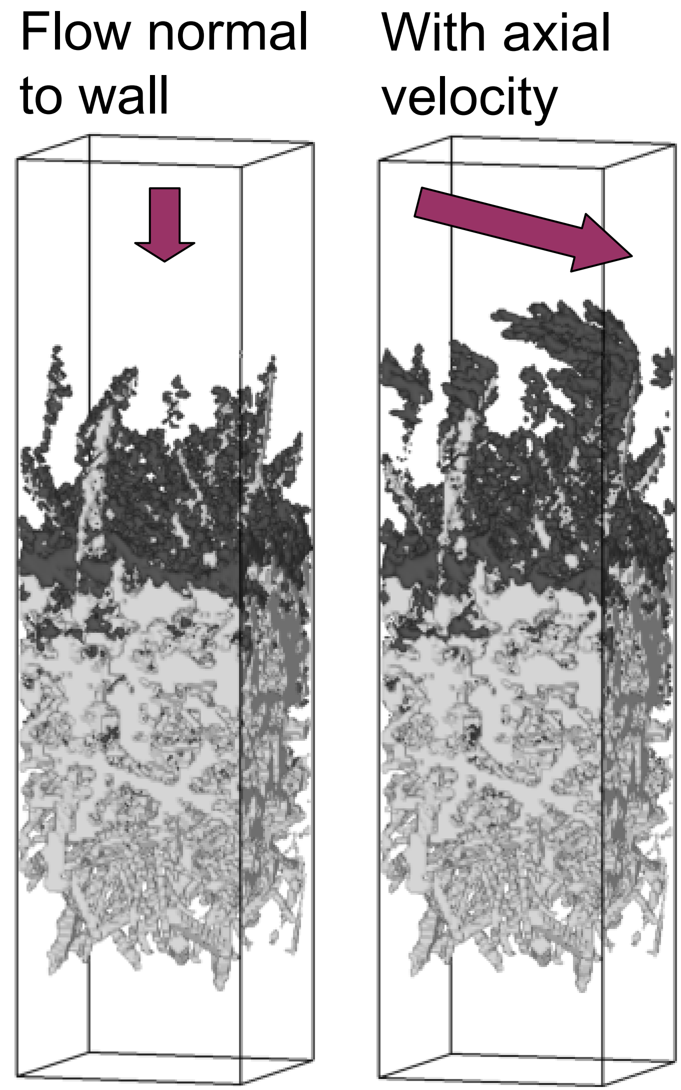
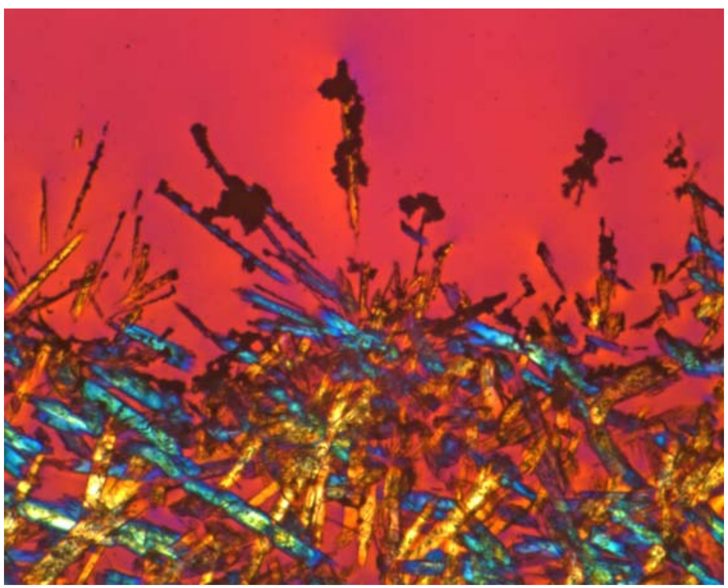
Deposits at needle tips due to axial flow

- Including an axial velocity component leads to preferential deposition at the tips of the tallest needles
- Less soot forced into the mouths of pores at the bases of needles could result in lower back-pressure
- Early deposits at the needle tips have also been observed during experiments



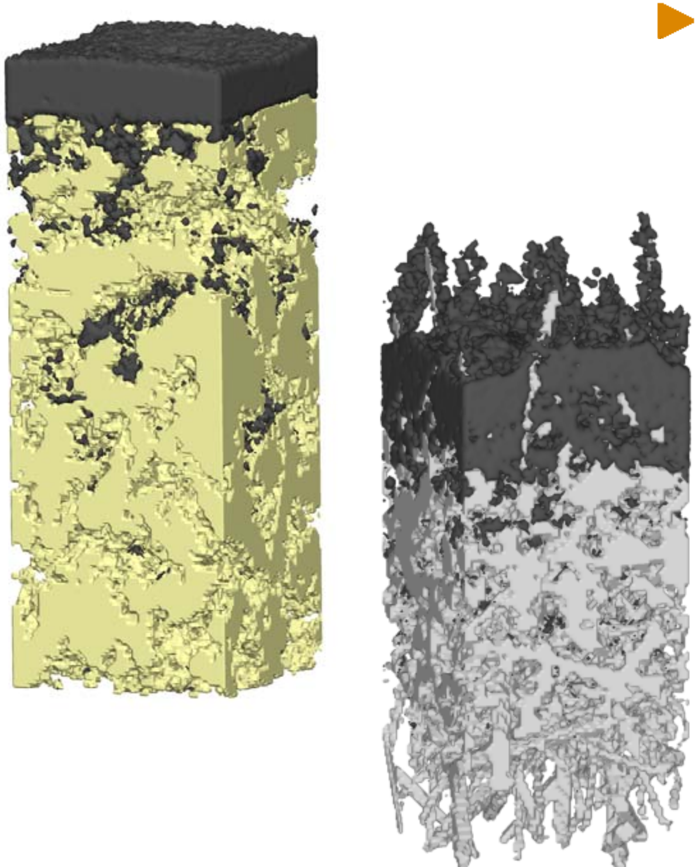
Filtration simulations



Thin section of a lightly loaded ACM filter wall

Initial passive regeneration simulations

- Uniform and infinitesimally thin layer of catalyst assumed
- NO_X oxidation kinetics for alumina-supported Pt used initially
  {S.S. Mulla et al. Journal of Catalysis 241 (2006) 389-399}
- Kinetics for advanced catalyst system on ACM is being incorporated
- Soot oxidation kinetics after Messerer et al. Carbon 44 (2006) 307-324



- Four cases for each substrate:
  1. No catalyst
  2. Catalyst distributed over entire filter wall
  3. Catalyst distributed over top quarter of wall
  4. Catalyst distributed over top eighth of wall

- Conditions:
  - Roughly 5.9 g soot/m^2 (5.3 g/L)
  - Regeneration subsequent to loading
  - Platinum loading: 120 g Pt/ft^3
  - 350°C
  - 550 ppm NO
  - 150 ppm NO_2
  - 10% O2

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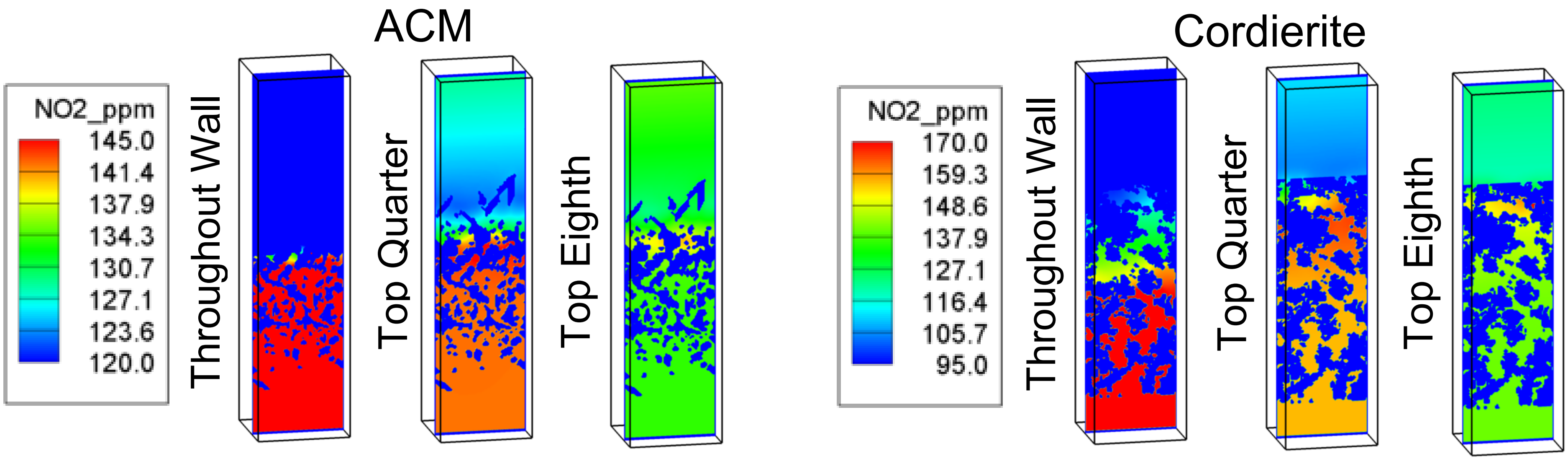
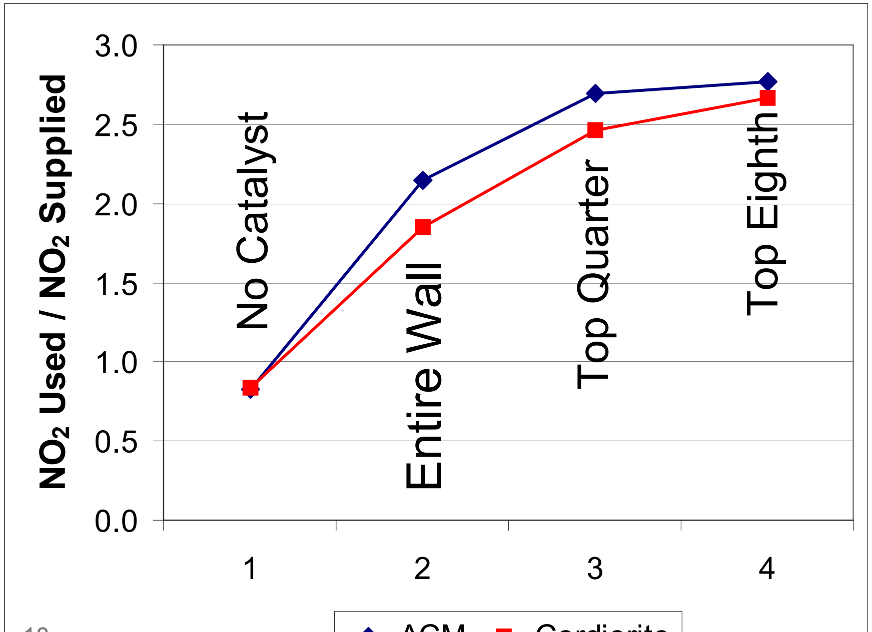
Effect of catalyst distribution





- Some back-diffusion of NO_2 from catalytic reactions within wall
- Wall nevertheless provides significant resistance to diffusion
- NO_X recycle promoted by placing catalyst close to soot
- Lower tortuosity of ACM may provide a small advantage for catalyst located within the wall

Summary

- Needle structures protruding from the filter wall surface appear to play an important role in the lower back-pressures observed during filtration with ACM
  - The needle structures tend to prevent complete blockage of pore mouths at their bases
  - The soot cake that forms among the ACM needles has a lower average density and higher permeability
- The effect of the protruding needles on total pressure drop is most apparent during the transition between depth and cake filtration
- Preferential collection of soot at needle tips due to axial flow may also tend to lower back-pressure during loading
- High surface area and relatively open structure of ACM may promote some filter regeneration mechanisms
- Strength of pore-scale modeling is in probing fundamental mechanisms and examining what-if cases

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