Development of a Sub-Grid Model of a Diesel Particulate Filter:

application of the lattice-Boltzmann technique

LDRD (an internally funded program at PNNL)

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Key Technical Hurdles – wall flow DPFs

- filter plugging
  - carbonaceous materials
  - ash deposits
  - face plugging
- thermal failures
  - cracking
  - melting
  - catalyst absorption into the bulk
- filtration performance
  - trapping effectiveness
  - sulfate particulate production
- size and cost
  - high soot ignition temperature – precious metals and/or enthalpy

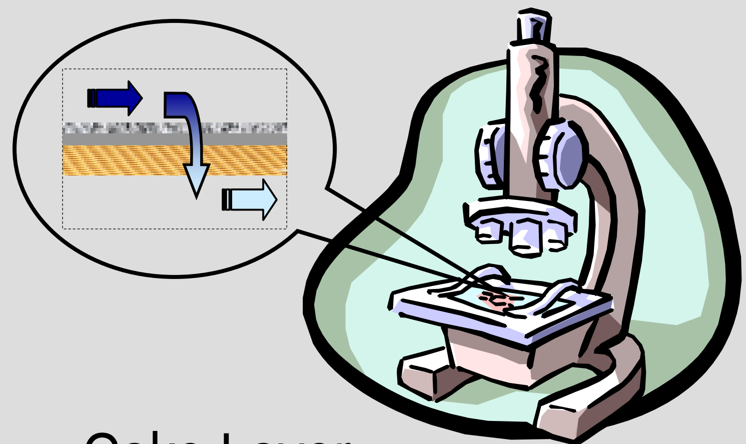
ref: Stover @ DEER 2001, Cummins Inc.

Representative Technical Questions - DPFs

- Why doesn't increasing pore diameter lead to reduced back pressure?
  - ref: Merkel et al., SAE 2001-01-0193
  - ref: Taoka et al., SAE 2001-01-0191
- Why does balance point temperature depend on soot loading level?
  - ref: Opris, DEER 2001
- What parameters affect soot cake permeability and packing density?
  - ref: Zhang et al., SAE 2002-01-1019
  - ref: Konstandopoulos et al., SAE 2000-01-1016
- How does morphology change with temperature and time?
  - ref: Yang et al., 3rd CLEERS Workshop, Oct 17, 2001
- What determines the level of soot loading at which a regeneration event can be made to be self-sustaining?
  - ref: Zelenka et al., SAE 2001-01-3199
- How does NO_2 diffusion from the catalyzed surface affect soot oxidation?

How “subgrid” Modeling Can Help

- filter plugging
  - carbonaceous materials
  - ash deposits
  - face plugging
- thermal failures
  - cracking
  - catalyst absorption into the bulk
  - melting
- filtration performance
  - trapping effectiveness
  - sulfate particulate production
- size and cost
  - high soot ignition temperature

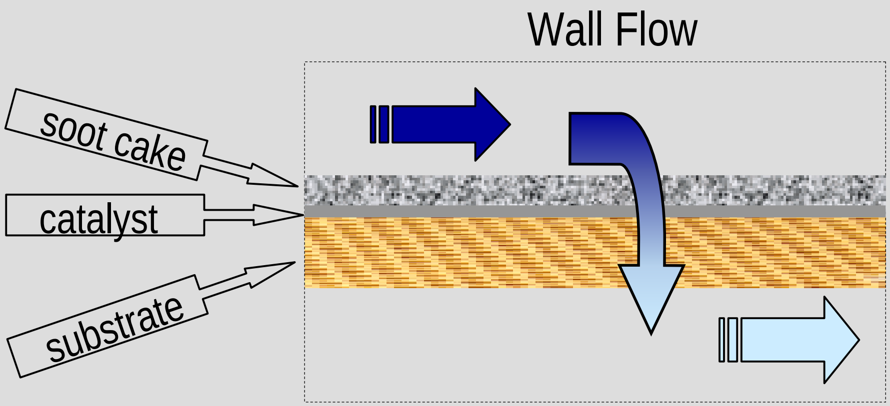


Cake Layer

- critical to filtration
- critical to back pressure
- critical to oxidation
- only ~15-50 microns thick
- involves heat & mass transfers, aerosol deposition, surface chemistry, catalysis...

Subgrid Physics – a complex problem

gas phase species
- O_2, O, O_3,...
- NO, NO_2, N_2O, N_2, HNO_3, HONO,...
- SO, SO_2, SO_3, H_2SO_4,...
- CO, CO_2 ,...
- H_2O, OH, H, H_2,...
- $C_aH_bO_cN_d$,...
- ...



solid phase compounds
- gamma-alumina
- sulphates, sulphites, ...
- cordierite
- Pt, Pd, Rd,...
- submicron carbonaceous particles
- phosphates, zinc, calcium, other ashes
- ...

processes
- bulk flow
- Darcy's law pressure
- permeation
- particle depositions
- diffusion
- surface reactions
- soot ignition
- oxidations/exotherms
- gas-solid interactions

reactions
- carbon and NO_2
- carbon and O_2
- microwave
- IR
- plasma
- others...

Computational Fluid Dynamics (CFD)

Begins with mass, momentum & energy balances

- a continuum conservation approach
- ie. draw a box and track convection, sources, and sinks

conserve mass ⟷

$$\frac{\partial(\rho u)}{\partial x} + \frac{\partial(\rho v)}{\partial y} + \frac{\partial(\rho w)}{\partial z} = 0$$

conserve momentum ⟷

$$\rho\left(\frac{\partial u}{\partial t} + u\frac{\partial u}{\partial x} + v\frac{\partial u}{\partial y} + w\frac{\partial u}{\partial z}\right) = \rho g_x - \frac{\partial p}{\partial x} + \mu\left(\frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} + \frac{\partial^2 u}{\partial z^2}\right)$$

conserve energy ⟷

$$\frac{\partial}{\partial t}\left[\rho\left(e+\frac{1}{2}v^2\right)\right] + \frac{\partial}{\partial x}\left[\rho u\left(e+\frac{1}{2}v^2\right)\right] + \frac{\partial}{\partial y}\left[\rho v\left(e+\frac{1}{2}v^2\right)\right] + \frac{\partial}{\partial z}\left[\rho w\left(e+\frac{1}{2}v^2\right)\right] =$$

$$k\left(\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2}\right) - \left(u\frac{\partial p}{\partial x} + v\frac{\partial p}{\partial y} + w\frac{\partial p}{\partial z}\right)$$

$$+\mu\left[u\frac{\partial^2 u}{\partial x^2} + \frac{\partial}{\partial x}\left(v\frac{\partial v}{\partial x} + w\frac{\partial w}{\partial x}\right) + v\frac{\partial^2 u}{\partial y^2} + \frac{\partial}{\partial y}\left(u\frac{\partial u}{\partial y} + w\frac{\partial w}{\partial y}\right) + w\frac{\partial^2 u}{\partial z^2} + \frac{\partial}{\partial z}\left(u\frac{\partial u}{\partial z} + v\frac{\partial v}{\partial z}\right)\right]$$

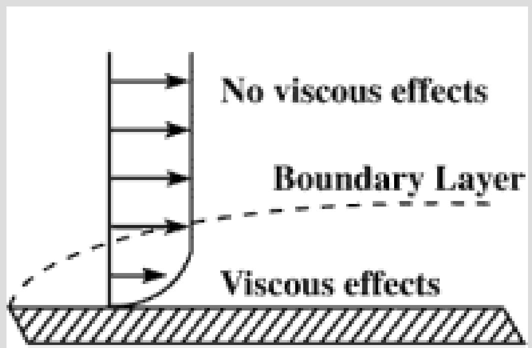
$$+2\mu\left[\left(\frac{\partial u}{\partial x}\right)^2 + \frac{\partial u}{\partial y}\frac{\partial v}{\partial x} + \left(\frac{\partial v}{\partial y}\right)^2 + \frac{\partial v}{\partial z}\frac{\partial w}{\partial y} + \left(\frac{\partial w}{\partial z}\right)^2 + \frac{\partial w}{\partial x}\frac{\partial u}{\partial z}\right] + \rho u g_x + \rho v g_y + \rho w g_z$$

Computational Fluid Dynamics (CFD)

The resulting equations are typically intractable

- boundary and initial conditions are killer
- only the simplest cases have explicit solutions

Blasius



$$ff'' + 2f''' = 0 \text{ with}$$

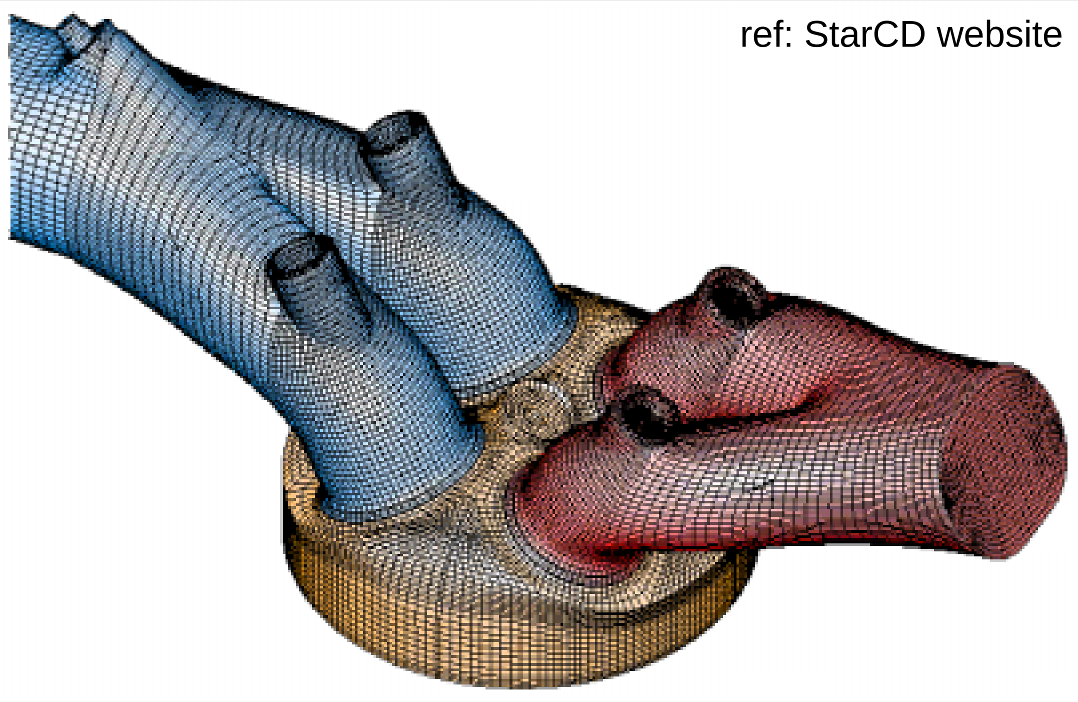
$$f = f' = 0 \text{ as } \quad = 0$$
$$f' \rightarrow 1 \text{ as } \eta \rightarrow \infty$$

$$u = Uf', \quad v = \frac{1}{2}\sqrt{\frac{U\nu}{x}}(\eta f' - f)$$

- numerical solutions partition the independant variables approximating the partial derivatives with solvable algebriac approximations

Computational Fluid Dynamics (CFD)

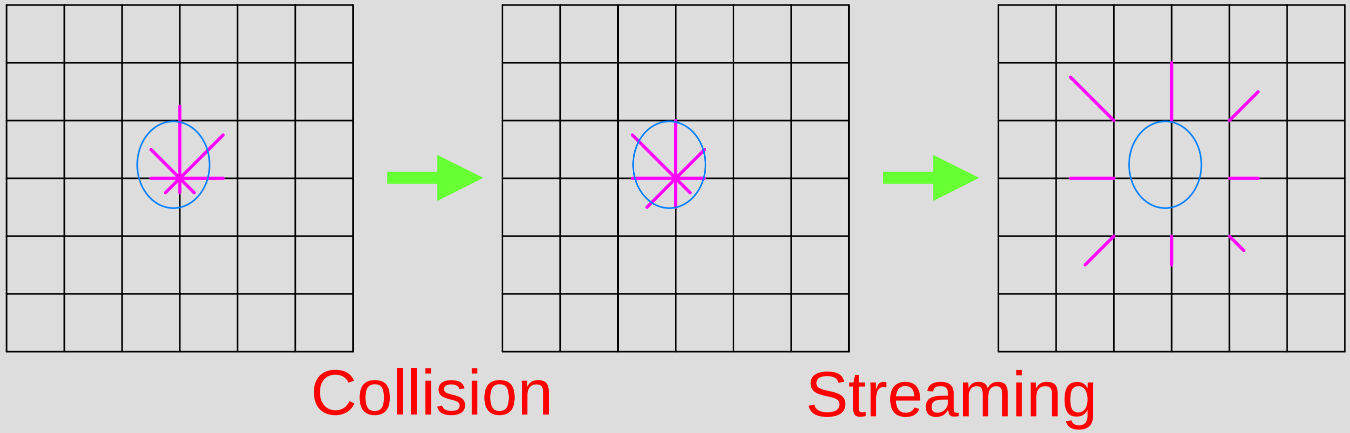
Complex geometries are difficult to grid, hence difficult to accurately model with CFD



Lattice-Boltzmann Techniques

Based on a Statistical Mechanics Approach

- the Boltzmann distribution function is the probability of a particle being at a specific velocity and location in time
- Boltzmann distribution function is discretized to a finite set of directions
- information from each lattice site is streamed to adjacent sites and relaxed toward equilibrium at each time step
- distributions are summed at each site to yield updated states



Why Use a Lattice-Boltzmann Technique?

- Lattice Boltzmann is second order accurate (same as CFD)
- Advantages
  - Easy to implement complex boundary conditions
  - CFD assumes isotropic pressure; LB full pressure tensor allows the incorporation of surface forces (surface tension, adsorption, etc.)
  - Global pressure solution not required, resulting in an inherently parallelizable algorithm (models consist of 10^6- 10^8 nodes)
  - Flexibility for incorporating new physics

Sub-grid Soot Filter Modeling - Components

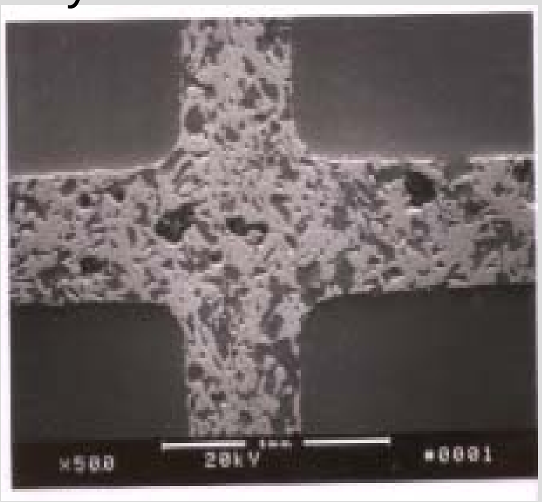
The following components are required for subgrid models:

- Physical Domain
- Representation of Soot in the Exhaust Stream
- Soot Adherence Characterization
- Soot Cake Layer Structural Properties
- Substrate Material Properties
- Catalytic Coating Characterization
- Reaction Mechanisms and kinetics
- Local Flow Field Representation (Heat and Mass x-fer)
- Exhaust Gas Temperature and Gas Phase Constituents

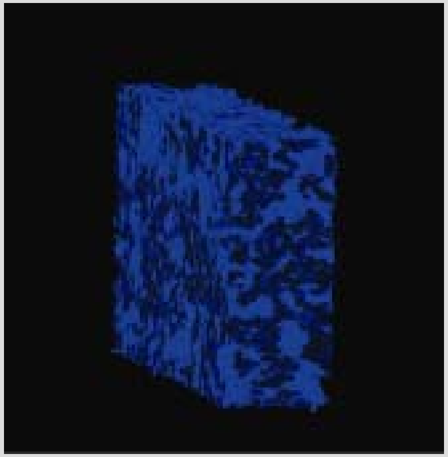
Physical Domain Characterization

Initial Domain: A highly porous cordierite diesel particulate filter substrate (e.g. Corning EX-80) which is a “tortuous path” structure with high local irregularities

Physical

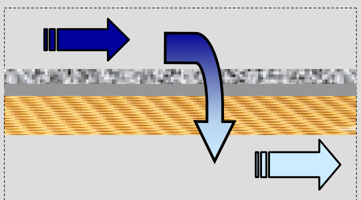


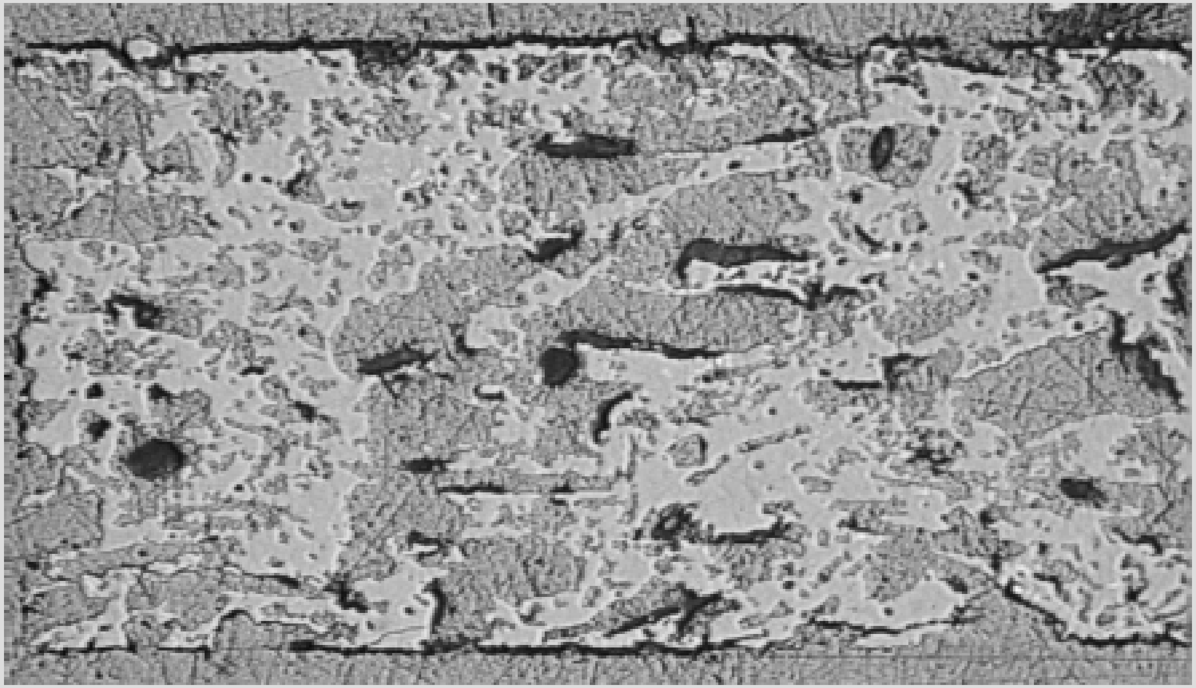
Computational



1. Sectioning and imaging

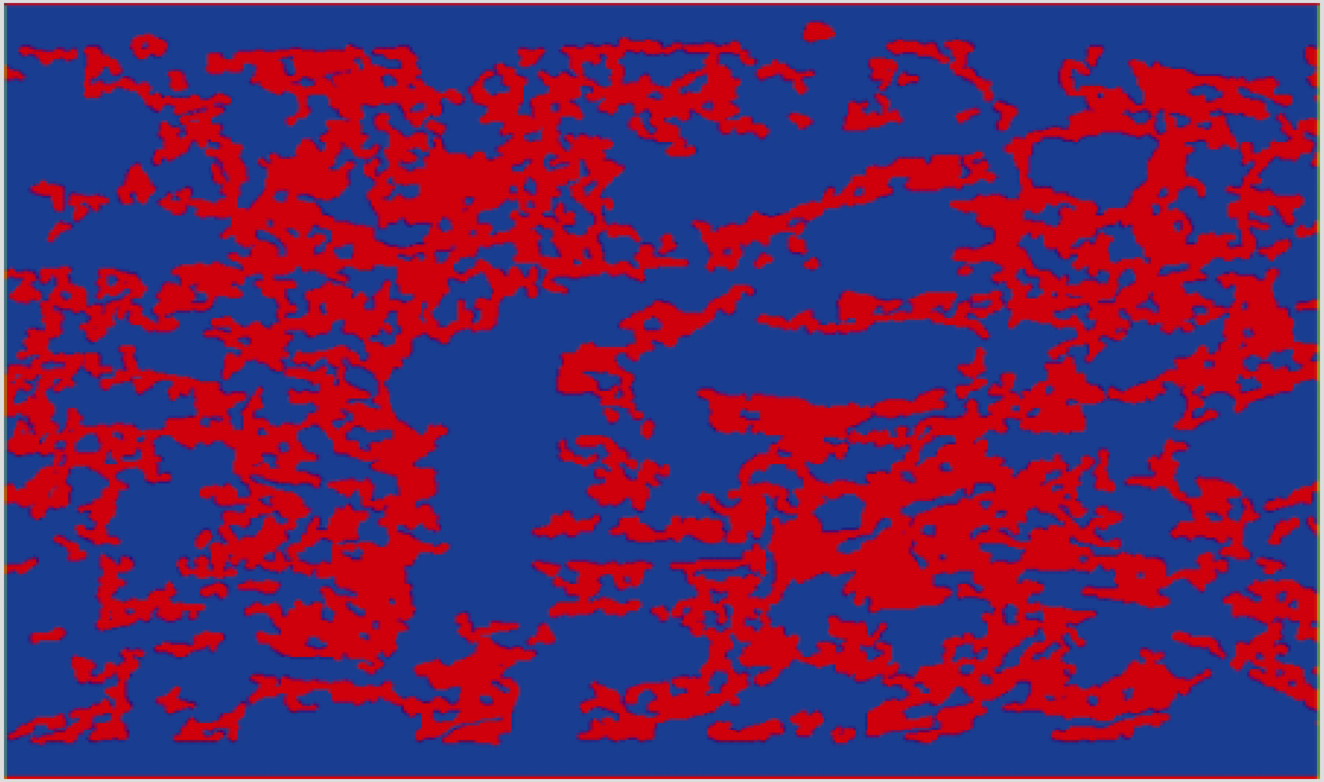
100 cpsi, Corning EX-80



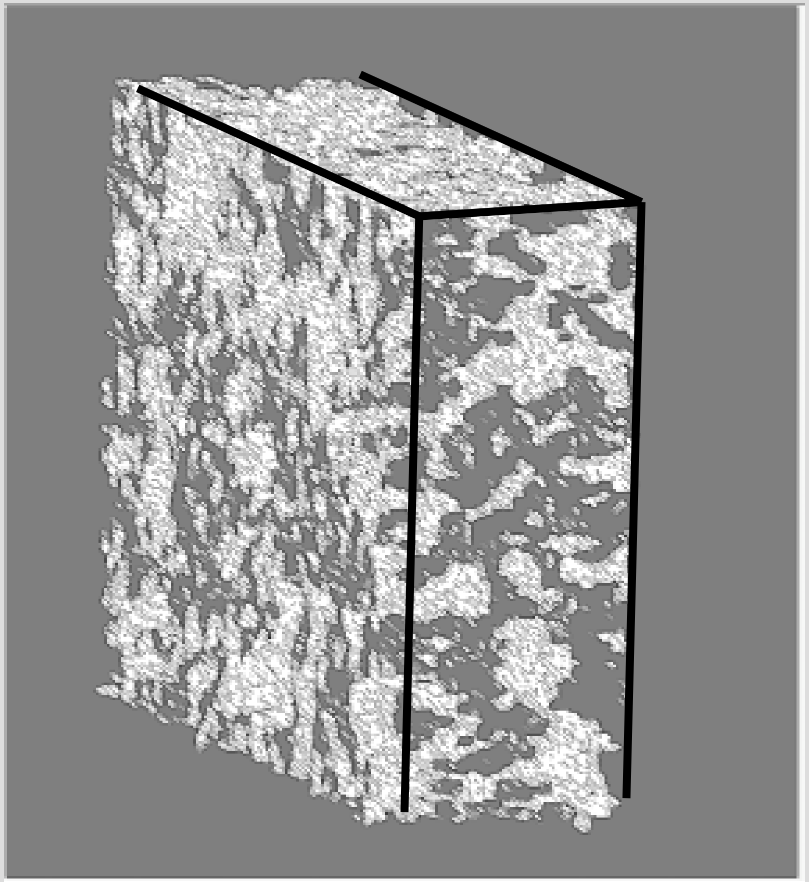


2. Image Digitization

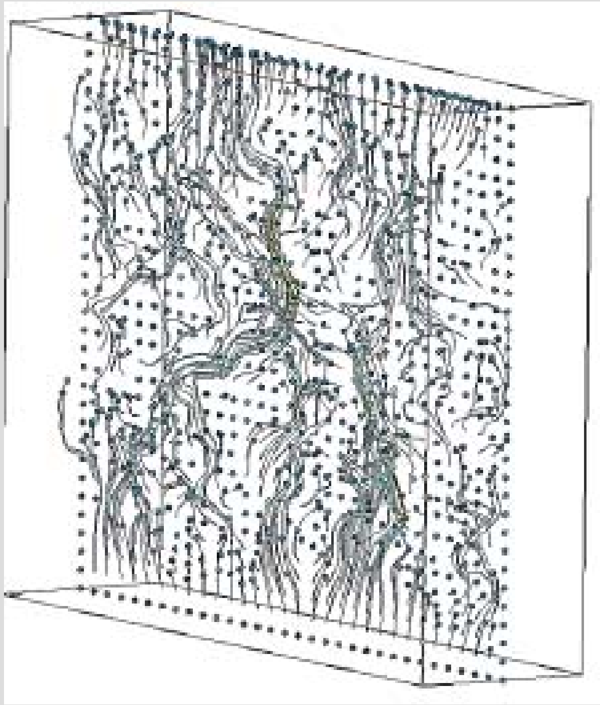
red – substrate, blue – void space

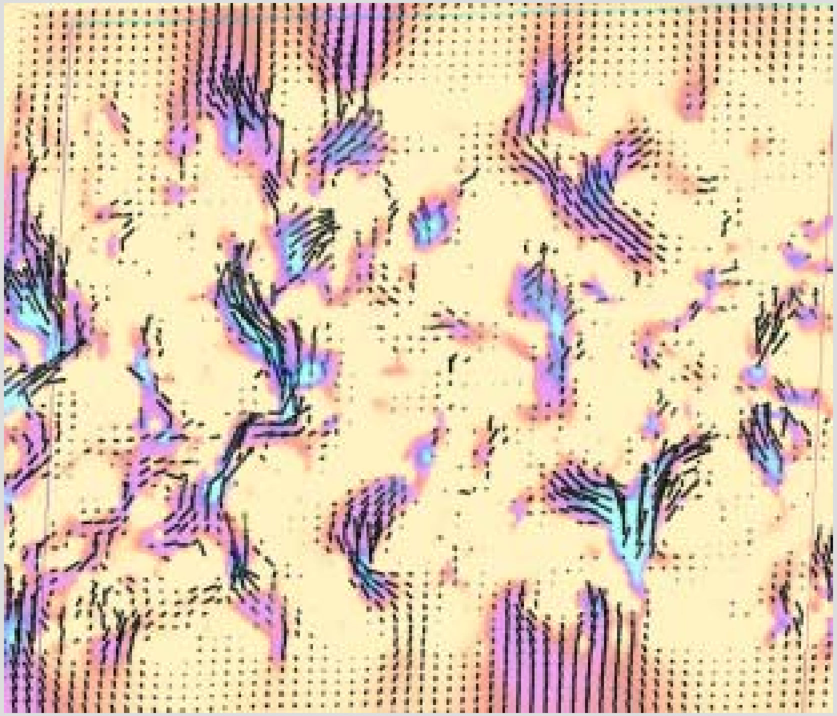


3. Interpolation and Reconstruction



4. Heat and Mass Transfer





5. Chemical Kinetics

Catalyst

- Net reactions $NO + {}^{1}/_{2}\, O_2 = NO_2$ and others
- Actual reactions involve surface structures

Gas phase reactions

- Mostly slow at exhaust temperatures without radicals from catalyst or plasma discharge

Soot oxidation

- Some possible net reactions:
  - $C(s) + 2NO_2 \Rightarrow CO_2 + 2NO$
  - $C(s) + NO \Rightarrow CO + NO$

6. Soot Oxidation

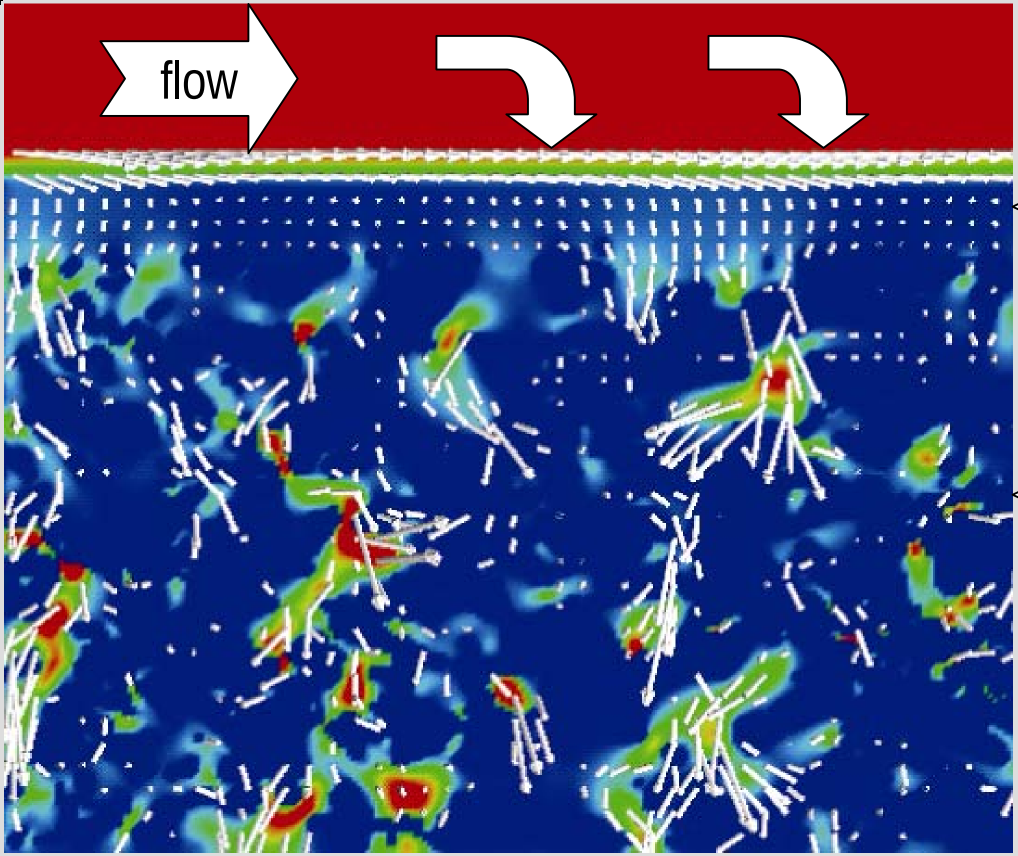
- Simplest case: no H_2O or SO_2
- Postulated model is analog of model of Kamm et al for C oxidation by O_3
- Our calculations used I, II.b, and II.c, with steady state parameters estimated from data

I.	$SS + NO_2 \Rightarrow SS.O + NO$
II.a	$SS.O + NO_2 \Rightarrow SS + NO + O_2$
II.b	$SS.O + NO_2 \Rightarrow SS' + CO_2 + NO$
II.c	$SS.O \Rightarrow SS' + CO$

7. Soot Cake Layer Approximation

- Assume a continuum
- Quasi-steady state
- Darcy law flow resistance
- 50 microns thick
- 5-10 micron pore penetration

Illustrative Example – local bottlenecks

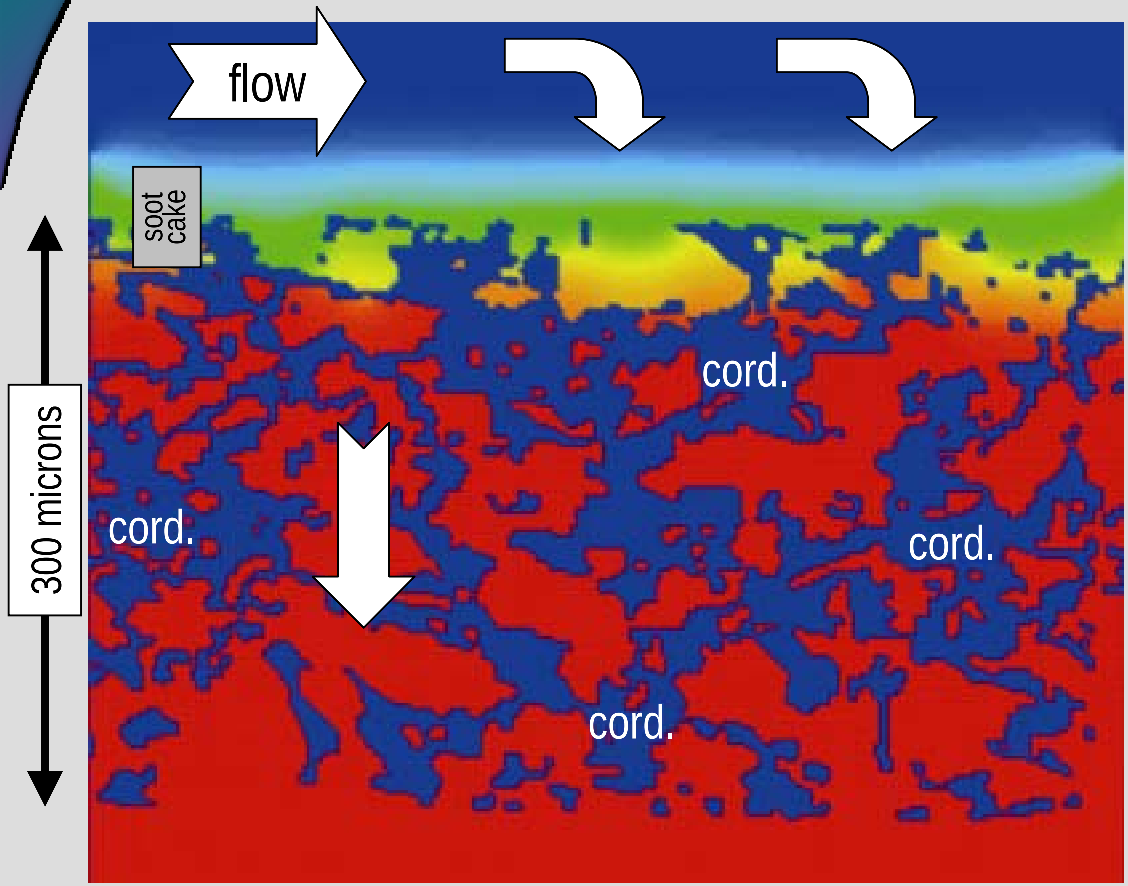


low gas velocities in and near the cake layer

high local gas velocities in few central bottlenecks

How does this structure impact backpressure? Can soot migrate to bottlenecks?

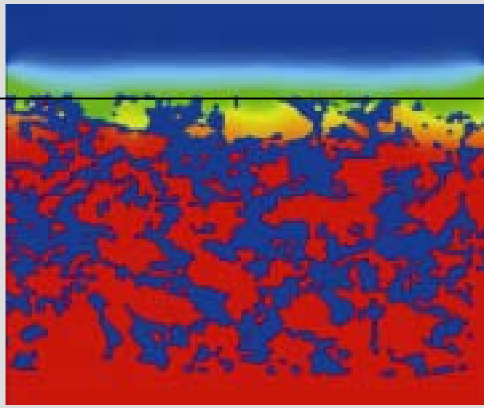
Illustrative Example – NO_2 back diffusion



<u>Conditions</u>

catalyzed surface (top 10 microns)
100ppm NO_2
900ppm NO
350 deg C
5 cm/sec face velocity

Illustrative Example – NO_2 “hot spots”

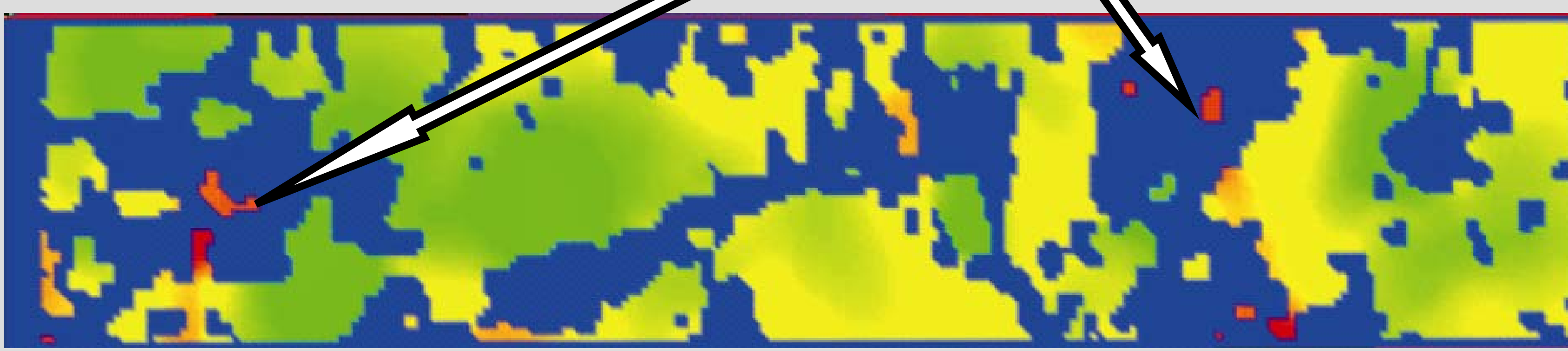


local “hot” spots (higher NO_2)

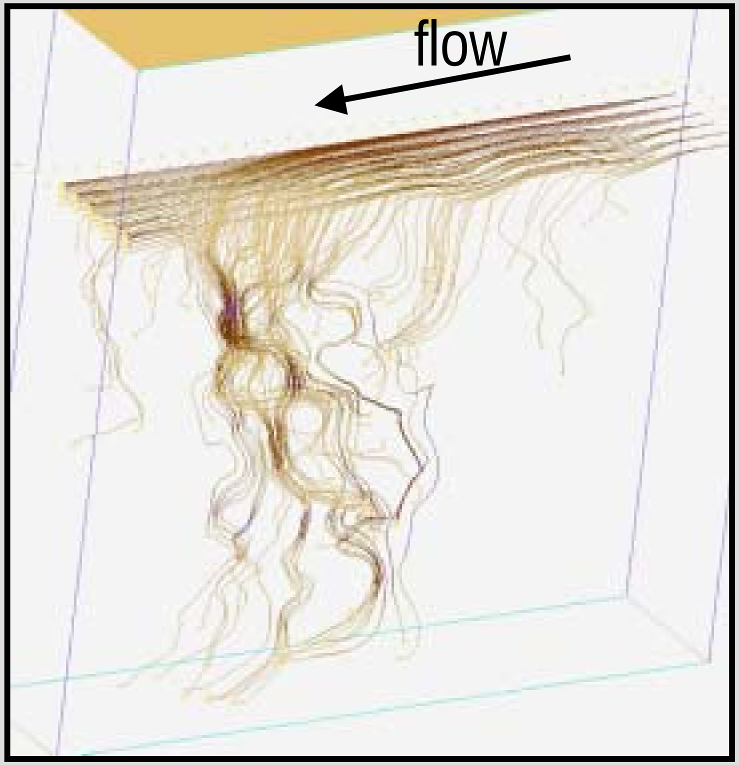
interplay between convection and diffusion

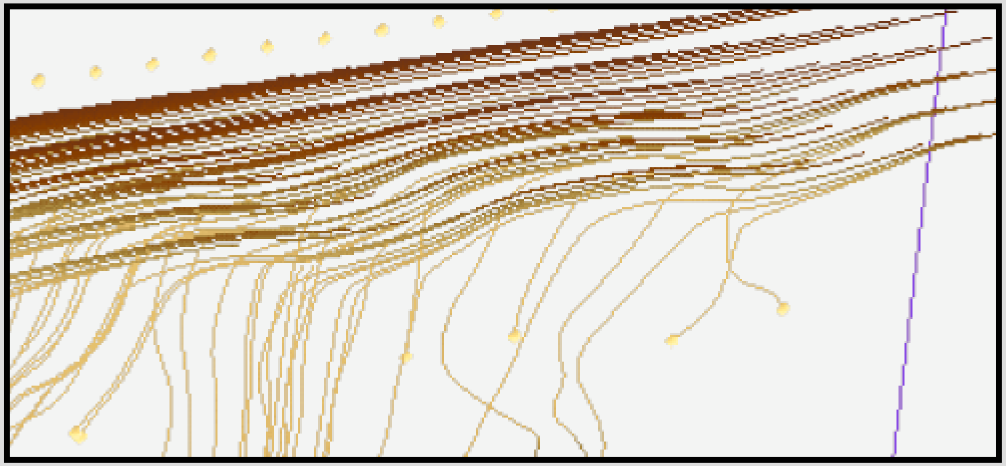
possible ignition points?

can these be tailored for improved performance?



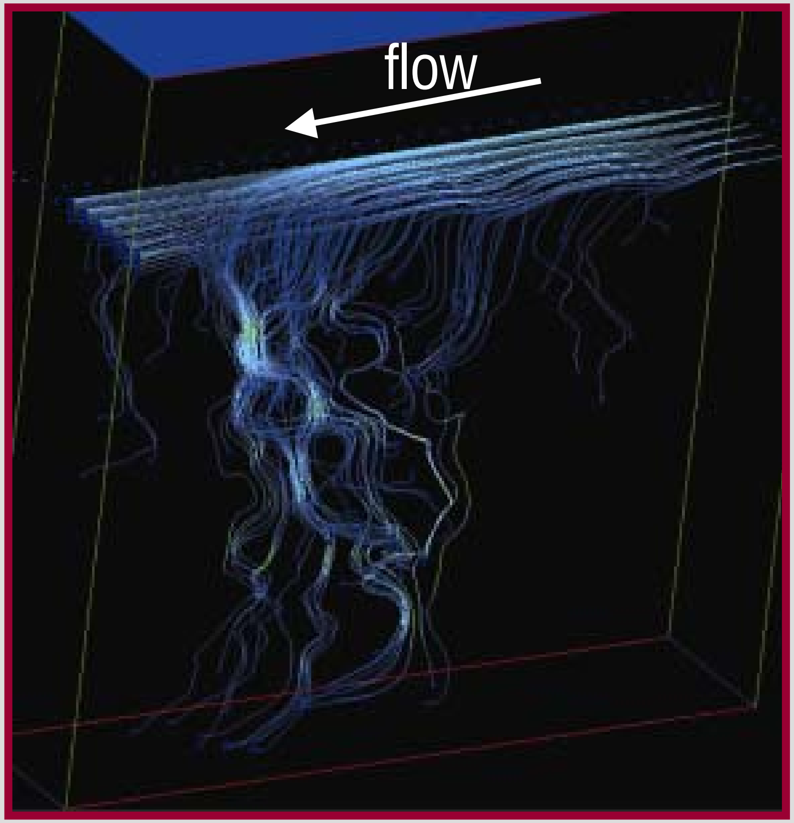
Illustrative Example – dynamic cake layer

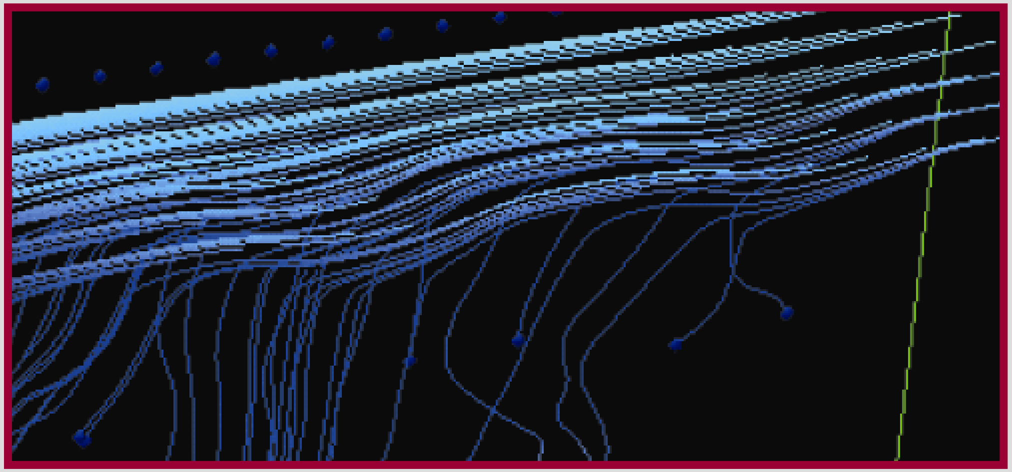




<u>Flow turns inward from troughs</u>
implies soot deposition in troughs?
implies cake layer thickness evolution?

Illustrative Example – dynamic cake layer





Flow turns inward from troughs
implies soot deposition in troughs?
implies cake layer thickness evolution?

Future

- LDRD will enter second year of funding in October
- Continue refining kinetics and soot deposition models
- Experimental validations
- sub-subgrid lattice-Boltzmann modeling of soot deposition during cake layer formation
- Next public report on progress will be at SAE congress in spring 2003
- All results and models will be incorporated into the CLEERS database
- Apply to Alternate Technologies
  - fibrous filtration...
  - NO_x aftertreatment...