

Lattice-Boltzmann Diesel Particulate Filter Sub-Grid Modeling – a progress report

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ABSTRACT

Aftertreatment modeling capabilities are an important part of the diesel engine manufacturer's efforts to meet the quickly approaching EPA 2007 heavy-duty emissions regulations. A critical, yet poorly understood, component of particulate filter modeling is the representation of the soot oxidation rate. This term directly influences most of the macroscopic phenomenon of interest, including filtration efficiency, heat transfer, back pressure, and filter regeneration. Intrinsic soot cake properties such as packing density, permeability and heat transfer coefficients remain inadequately characterized (1). The work reported in this paper involves subgrid modeling techniques which may prove useful in resolving these inadequacies. The technique involves the use of a lattice Boltzmann modeling approach. This approach resolves length scales which are orders of magnitude below those typical of a standard computational fluid dynamics (CFD) representation of an aftertreatment device. The improved resolution may allow for the characterization of functionality not previously reported in the literature.

This paper presents the first status report of this multiyear project. Descriptions of the modeling technique, the initial kinetics, and the development of the computational domain are provided. In addition, preliminary sample exercises are discussed in order to illustrate how the final model, once refined and validated, may be applied in practice.

INTRODUCTION

The United States Environmental Protection Agency's 2007 heavy duty diesel engine exhaust emissions standards mandate a 90% reduction in particulate matter over current levels. The majority opinion in the diesel industry is that these standards are, by implication, mandating exhaust particulate filtration for diesel

engines (2). Unfortunately, there is no commercially viable filtration technology which currently exists that can be universally applied to all on-highway heavy duty diesel engines without regard to duty cycle (3). Several key technical hurdles that must be overcome in order for diesel particulate filters to become practical include: filter plugging, thermal failures, size, cost, filtration performance, deactivation and durability.

The resolution of these technical issues requires a detailed system level understanding of the interaction between the diesel engine's exhaust characteristics and the filtration device. Attention must be given to all possible application duty cycles and ambient conditions. Modeling can greatly expedite this process. In order to be effective, three specific modeling tasks must be accomplished. First, detailed combustion and air handling models are needed in order to understand exhaust gas temperatures and constituents. Second, exhaust system models (including a characterization of individual installation requirements) are required for heat and mass transfer effects. This would also incorporate any sensors and/or actuators within the exhaust system. Third, the individual aftertreatment device performance must be adequately characterized. Only then can simulations effectively be produced to help aid in the understanding and prediction of filtration effectiveness, durability, and fuel economy impacts.

These system models can be used to optimize the engine control strategies, to help determine the physical dimensions of the catalyst system and to simulate numerous real world applications which may be prohibitively expensive to field test individually. The detailed component models can be used to investigate novel new filtration techniques or to optimize existing substrate geometries for soot oxidation or soot capacity.

Regardless of the specific soot filtration or system performance question being pursued, a critical key

component of these models is the representation of the soot oxidation rate (4, 5). This term directly influences most of the macroscopic phenomenon of interest including filtration efficiency, heat transfer, back pressure, and optimal substrate geometry; ref. Figure 1.

One key technical challenge in characterizing soot oxidation rates relates to the fact that particulate filter soot regeneration remains a poorly understood phenomenon, especially at the length scales of the soot cake layer. Current subgrid models use gross simplifications to represent the carbonaceous soot oxidation rates. All phenomenon which occur on the finest length scales (approx. <100 microns) are typically lumped together into a single Arrhenius-type equation with curve fitted pre-exponential and activation energy terms (4, 5). Furthermore, intrinsic soot cake properties such as packing density, permeability and heat transfer coefficients are poorly understood. Improvements in these subgrid model formulations will improve the ability of engineers to model the filter on a device scale leading to better system optimization capabilities and improved design tools.

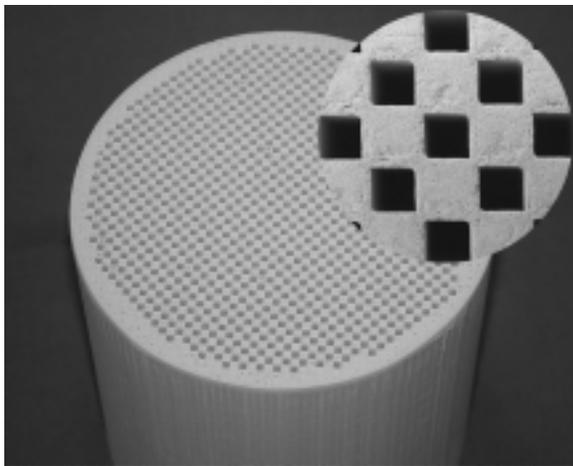


Figure 1 "cordierite diesel soot filter"

This paper reports on a technique which may assist in improved subgrid representations of the local oxidation reactions via detailed subgrid modeling. This technique involves the use of a lattice Boltzmann modeling approach. This approach resolves length scales which are orders of magnitude below those typical of a standard Computational Fluid Dynamics (CFD) representation of an aftertreatment device {*note: theoretically, CFD techniques can be used down to these smaller length scales. However, for reason discussed in the next section, the geometry of the pore structure makes this approach impractical*}. This improved resolution allows for the characterization of functionality not previously employed in the CFD analysis.

THE LATTICE-BOLTZMAN TECHNIQUE

The basic conservation equations (continuity, momentum and energy) which govern fluid dynamics are typically formulated as a set of coupled, nonlinear partial differential equations. These equations have no general closed form analytical solution except in the case of very trivial initial and boundary conditions (e.g. the Blasius solution for a flat plate). These equations are often referred to collectively as the Navier-Stokes equations - given the additional assumptions that Newton's viscosity law and Fourier's conduction law are employed (6). Due to the intractability of these equations, analytical solutions rarely exist for the practical problems in engineering. Therefore, computational techniques must be employed.

Conventional computational fluid dynamics methods involve the discretization of the Navier-Stokes equations using a finite volume or finite element formulation. In other words, the partial differentials are approximated locally over an "element" as algebraic differences. Given the size and shape of the element and the form of the algebraic approximation, varying degrees of accuracy can be obtained. However, as a general rule, the solution is governed largely by the quality of the computational mesh describing the physical domain. Highly intricate and complex geometries, such as the pores in a cordierite soot filter substrate, are very difficult to grid accurately and efficiently.

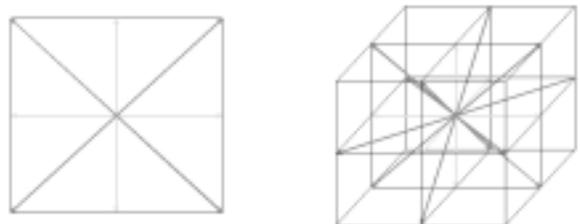


Figure 2 "Lattice displacement vectors for two- and three-dimensional systems"

In addition, the solution procedure typically involves solving for pressure throughout the entire domain at each time step to satisfy continuity. This pressure solution does not scale linearly on a parallel computer, limiting the maximum number of nodes that can be used to resolve a geometry. In addition, boundary conditions are difficult to implement for complex geometries such as porous media. For all these reasons, traditional CFD approaches have typically not resolved the fine scale structures in diesel particulate filters.

Lattice-Boltzmann computation is a powerful alternate to classical computational fluid dynamics. The classical techniques use finite elements or finite volume formulations. The lattice Boltzmann method takes a different approach to modeling fluid systems than conventional solvers (7, 8, 9). The spatial domain is discretized into a finite number of lattice sites, where

each lattice site has values for density, pressure, flow, etc. The single-particle distribution function, which describes the probability of a particle traveling along a particular direction and speed, is discretized to form a finite set of displacement vectors connecting each lattice site to adjacent sites, ref. Figure 2. Similar distributions are also created for energy and each chemical species.

The lattice Boltzmann equation describes the evolution of the discretized particle distribution function, $f_i(\mathbf{x},t)$, along direction i as a function of time. The new time distribution function is given by the equation

$$f_i(x + \Delta te_i, t + \Delta t) - f_i(x, t) = -\frac{1}{\tau} [f_i(x, t) - f_i^{eq}(x, t)]$$

where τ is a linear relaxation parameter and f^{eq} is the local equilibrium distribution. The local equilibrium is expressed in the form of a quadratic expansion of the Maxwellian distribution

$$f_0^{eq} = w_0 [A_0 + D_0 u^2]$$

$$f_i^{eq} = w_i [A + B(e_i \cdot u) + C(e_i \cdot u)^2 + Du^2]$$

where the coefficients are defined as

$$A_0 = \rho - 6A; \quad D_0 = -\frac{\rho}{c^2}$$

$$A = \frac{\rho}{3c^2}; \quad B = \frac{\rho}{3c^2}; \quad C = \frac{\rho}{2c^4}; \quad D = -\frac{\rho}{6c^2}$$

and c is the reference lattice speed, $c = \Delta x / \Delta t$. The weight coefficients, w_i for a three-dimensional system are

$$w_i = \begin{cases} \frac{1}{3} & i = 0 \\ \frac{1}{18} & i = 1-6 \\ \frac{1}{36} & i = 7-18 \end{cases}$$

The linear relaxation parameter, τ , is related to the kinematic viscosity by the expression

$$\nu = (\tau - 0.5)\Delta t c / 3$$

Solution methods consist of a streaming stage and a collision stage for each time step. During the streaming stage, each lattice site transmits distribution information with adjacent neighbor sites. The incoming information is then relaxed toward an equilibrium distribution, which is determined by local conditions. The new time pressure and velocity values are determined using the expressions

$$p = \frac{c}{3} \sum_i f_i$$

$$\rho u = \sum_i f_i e_i$$

This procedure is repeated for the specified number of time steps until a steady-state solution is obtained. The result is a second-order solution to the Navier-Stokes and continuity equations

$$\rho \frac{\partial u}{\partial t} + \rho u \cdot \nabla u = -\nabla p + \rho \nu \nabla^2 u$$

$$\nabla \cdot (\rho u) = 0$$

A similar procedure is followed for the solution of the energy and chemical species transport equations.

The advantages of this procedure center on the fact that no global solution methods are required for determining field variables. Each lattice site only requires information from adjacent sites. This makes the lattice Boltzmann method inherently parallelizable and makes it possible to simulate complex geometries in great detail. Pore scale geometries consisting of millions of lattice nodes are typical. Lattice-Boltzmann is well suited to compute flows with elaborate boundaries, such as flows through the pores of a ceramic diesel particulate wall filter. Lattice-Boltzmann can also treat phenomena on boundaries, such as chemical reactions at the surfaces of catalysts or soot particles, in addition to chemical reactions in bulk.

CONSTRUCTION OF THE PHYSICAL DOMAIN

In order to obtain meaningful prototypic modeling results using lattice-Boltzmann, the physical domain used in simulations must be as accurately represented as possible. Therefore, a wall-flow cordierite diesel particulate filter was selected, representing the most common filter media currently utilized by the aftertreatment industry. Corning EX-80 soot filters measuring 14.3 cm diameter and 15.2 cm long were procured, analyzed, and the geometry of the filter wall pore-structure digitized for use in defining of the physical domain and boundary conditions.

The development of the lattice Boltzmann model was a non-trivial matter due to the intricate nature of the physical domain. The EX-80 filter has an extreme tortuous-path flow profile. The substrate has a high fraction of void space, is three-dimensional in its structure, with a wide pore-size distribution. Since the accuracy of the modeling results depend on the quality and accuracy of the physical domain representation, detailed three-dimensional geometries of the porous soot filter material were required.

A procedure was developed for defining and digitizing the soot filter geometry from a series of micrographs. The EX-80 filter samples were sectioned in the three principle axis (normal to flow direction, longitudinal filter channel wall, and cross section of the filter wall), and approximately 1 cm by 1 cm specimens of each axis were potted in an acrylic epoxy microscope mount. The samples were then ground to the filter wall surface and polished flat, using a series of mechanical grinding and polishing techniques. The final polishing step was with a water-based colloidal suspension of 1-micron diamond polishing compound.

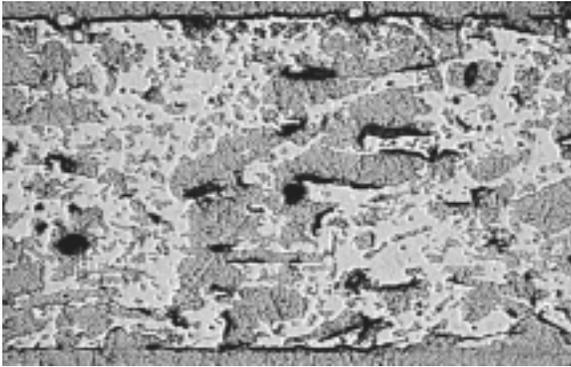


Figure 3 "micrograph of EX-80 substrate wall cross-section"

Optical microscopy images were taken and digitized for each sample, a shown in Figure 3. After the first round of images were taken all of the samples were then repolished with a 3-micron diamond and water colloidal media using a Buehler Automet[®] automatic polishing station. The subsequent repolishing step was performed for 60 seconds in order to remove approximately 6-microns of material from the filter surface. Additional images of each sample were then taken. This procedure of polishing and imaging was repeated 25 times and produced a series of 25 surface images spaced 6-microns apart of the porous filter wall structure.

Upon completion of the microscopy work, the 25 images were processed with image analysis software and converted into discrete digital representations of the solid filter material and void space, as shown in Figure 4. Each image, or layer, was then combined with a custom written interpolation code in order to fill in the domain in between each layer. Ultimately this process created a continuous three-dimensional space representation of the actual filter geometry, shown in Figure 5.

Sample sets representing the three-dimensional structure were taken from multiple locations in the EX-80 filter for comparison of the homogeneity of the wall structure and pore-size distribution on a global scale, and to provide multiple domain sets for flow field analysis. The digitized three-dimensional physical domain was then meshed and used in the lattice-Boltzmann model to define the filter geometry and

boundary conditions.

The lattice Boltzmann model of the filter material used in these simulations consisted of 240x240x60 lattice nodes with approximately 3 micron resolution. Each lattice node has an associated velocity vector, pressure, temperature and molar concentration for each chemical species. This level of resolution allows the detailed three-dimensional representation of flow and field variables within the pore structure of the filter material.

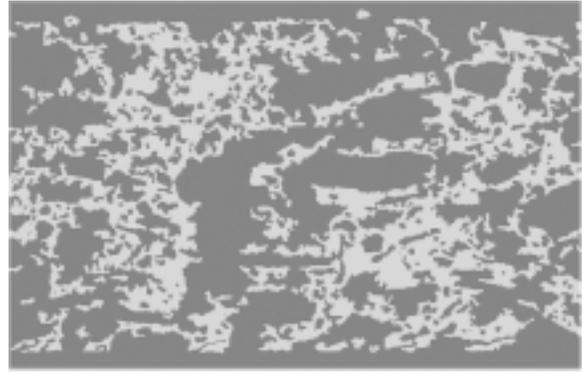
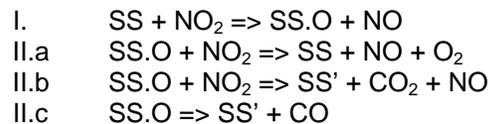


Figure 4 "digitized image of micrograph from figure 3"

CHEMICAL KINETICS/SOOT OXIDATION

A preliminary model for soot oxidation was implemented for the purpose of running sample exercises (described in a later section of this paper). This model assumes the soot is oxidized by NO₂. It was developed in analogy to a model in which O₃ was the oxidant, as proposed by Kamm et al. (10).

The reactions proposed as occurring at a surface site (SS) on the soot cake are:



Models for H₂O, OH and SO₂ effects, including site poisoning and passivation, are to be added to this model at a later date. For the sample exercises calculations, reactions I, II.b, and II.c were used, with rates inferred from experiments reported in the literature, assuming steady state conditions. Note that in this model, emission of CO is a spontaneous thermal process after the first reaction (I) of NO₂ with the surface. CO₂ emission, by contrast, requires a continuing NO₂ flux. For each NO given off, according to this model, an NO₂ has disappeared. The current model contains these and other testable hypotheses, and extensions will undoubtedly be required as new data is acquired.

A theme of the proposed reactions is that they all plausibly give negative free energy changes, and require no intermediate states or reaction products with high heats of formation. The verification of the existence of a

$R^A C(R^B)=O$ or $R^A C(R^B)(R^C)-O^*$ structure on the surface from its vibrational absorption bands for two or three of these cases (NO_2 , O_3 , SO_3) would be informative, indicating that the analogy to the model of Kamm et al. (10) has some validity.

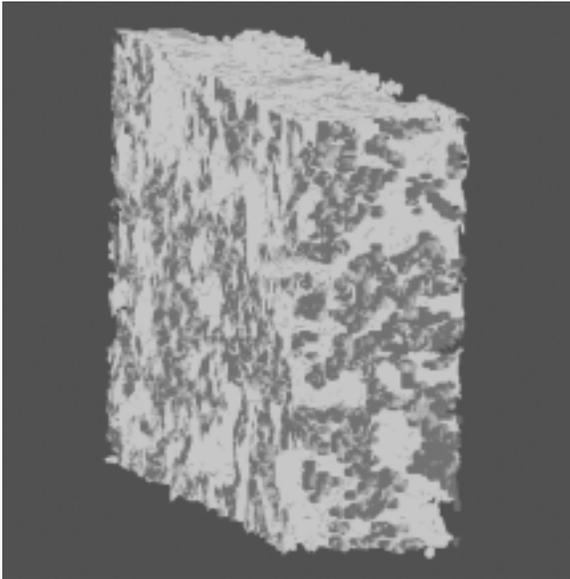
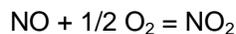


Figure 5 “digitally reconstructed physical domain”

Rate parameters for this model were determined from the changes in NO , NO_2 , CO , and CO_2 concentrations in flow of an initial NO_2 - NO mix through a diesel soot sample at temperatures from 100 to 780 C, as reported by Choi et al. (11). A steady state value of the surface site concentrations was assumed.

Models for catalysis were even more simplified, with rates for the net reaction



occurring on pure platinum inferred from rates in the literature for Pt supported on SiO_2 . Assuming that the O_2 came from the catalyst surface and that the O_2 availability on the surface is not the rate-limiting process a temperature-dependent probability of conversion of an incident NO to NO_2 on platinum was determined from the data of Liu et al. (12) for use in our simulations. The rate of conversion of NO to NO_2 per unit area of catalyst surface is thus $f(NO \rightarrow NO_2) * F(NO)$, where $F(NO)$ is the incident NO flux, and $f(NO \rightarrow NO_2)$ is the conversion probability. For our simulations, the platinum on or near the surface was assumed equivalent to a mono-layer of platinum for reaction rate.

SOOT CAKE REPRESENTATION

For the sample exercises the top surface of the filter material was assumed to have a layer of soot cake already deposited. The size of soot particles in the cake can range from 10 nm up to one micron, which is less than the node resolution used to represent the filter

material. Therefore, the soot cake was represented as a continuum with an associated Darcy flow resistance. The soot cake was assumed to be 50 microns thick and overlapping the filter material so that there is a 5-10 micron penetration of soot cake into the filter. The cake was assumed to have a uniform solid density and surface area available for chemical reaction.

The next phase of research will focus on refining the soot cake model, including the transient evolution of the soot density profile due to deposition, loss due to oxidation and cake consolidation.

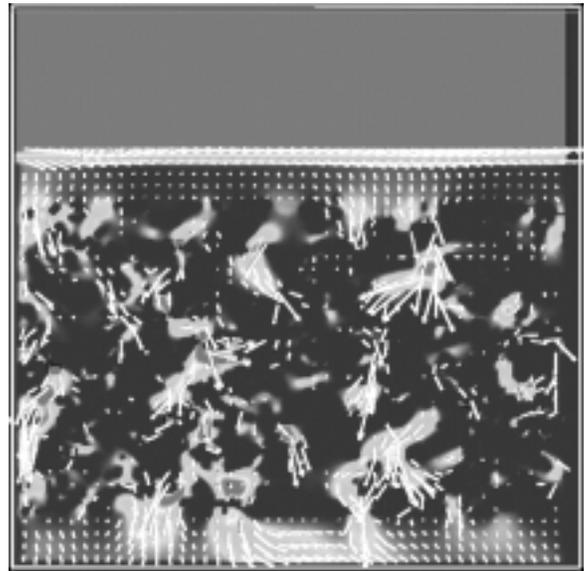


Figure 6 “flow velocity vectors – test Case 1”

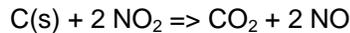
SAMPLE EXERCISES

As a prototypic question to be addressed with Lattice-Boltzmann analysis, we considered flow through the configuration described in the earlier sections with two different assumptions for the soot layer configuration. Case 1 assumed a uniform soot cake layer. Case 2 had a soot layer thickness adjusted to be proportional to the perpendicular flow velocity from Case 1. These cases were run simply to illustrate the type of questions which can be addressed with the lattice-Boltzmann technique. It should be emphasized that these results have not been experimentally validated at this time.

CASE 1

The motivation for the Case 1 study is the desire for a better understanding of the phenomenon known as “ NO recycling”. One approach to filter regeneration is to have NO -to- NO_2 conversion occur in the midst of deposited soot, so that the produced NO_2 is quickly converted back to NO by useful soot-oxidizing reactions, minimizing back-conversions by less productive means {with increasing temperature in a gas containing a significant amount of O_2 , NO_2 becomes decreasingly preferred compared to NO . NO_2 is more effective for

soot oxidation, and is probably the preferred form for catalytic reduction to N_2 . The reaction rate of NO_2 with soot increases with increasing temperature, but, as might be expected, the rates of other back reactions which convert NO_2 back to NO also increase with temperature. One net reaction for one sequence of NO_2 reactions on soot is



In which each NO_2 gets reconverted to NO while consuming half a solid C. "Recycling" the NO by repeated steps of NO -to- NO_2 catalytic conversion followed by C-oxidizing NO_2 -to- NO conversions offers some promise for making filter regeneration more effective.

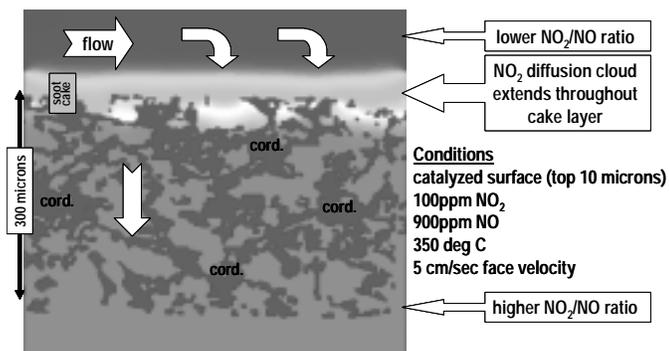


Figure 7 "NO₂ concentration gradients – test Case 1"

Experiments reported in the literature have explored the requirements for closeness of contact between catalysts. The sample exercise reported here explores such issues as the proximity, flow direction, and residence requirements for one such configuration in which the catalyst has its effect by oxidizing NO to NO_2 .

The conditions modeled in Case 1 included 100ppm of NO_2 and 900ppm of NO in the feed gas. The temperature of the gases, and the substrate far field were held at 350deg C. The face velocity of the exhaust gases were held at 5 cm/sec. Given the extensive gradients of temperature and flow within a complete soot filter, it is not possible to exactly correlate these local conditions on the subgrid with inlet exhaust conditions in an actual engine experiment. However, these values were felt to correlate with a typically sized soot filter device operating at a modest load condition on a current diesel engine configuration.

Figure 6 is an illustration of the flow field resulting from executing Case 1. It depicts flow velocity vectors at a sampling of representative lattice nodes. Perhaps the most intriguing aspect of this result is the appearance of "internal bottlenecks". These are locations of high local flow velocity within the substrate structure. In fact, the flow through the surface of the substrate near the cake layer is quite uniform. The highest flow velocities appear to be associated with regions of pore intersection. Given that the cake layer is quite thin and located near the

inner surface of the substrate, these pore intersections are not found in this region. The appearance of these internal bottlenecks leads to some interesting postulations. For example, can it be that these internal bottlenecks can become the primary location for high back pressure in an operating soot filter where the soot cake has lost some of its structural integrity and pieces of the cake have migrated to these local bottlenecks. This may help explain the curious results reported by Merkel et al. (13) in which larger pore diameter substrates actually had higher backpressure. Taking this hypothesis one step further, could a structure be created where these local bottlenecks did not occur? and would the resulting structure be more resistant to plugging? Again, these types of questions are presented only to illustrate the utility of subgrid modeling. No firm conclusions can be drawn till experimental validation has led credibility to the modeling.

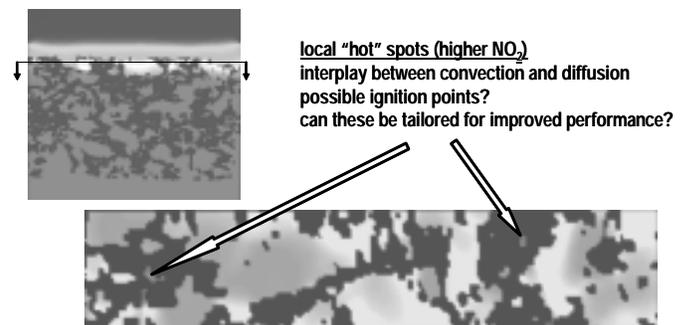


Figure 8 "horizontal section from figure 7"

Another interesting result from the simulations of Case 1 involves the interaction between NO_2 diffusion from the catalyst sites and the general flow convection tending to carry away the NO_2 from the soot cake region. Figure 7 illustrates the concentration gradients of NO_2 . As can be seen in the figure, NO_2 is produced via reactions on the surface and does, in fact, back diffuse into the soot cake layer. Clearly, under these operating conditions diffusion can overcome the convective effects. Figure 8 illustrates a cross section from figure 7 directly at the surface of the substrate. It is quite apparent that there exists substantial local "hotspots". These hotspots show areas of high NO_2 and are related to regions along the surface where the interplay between diffusion and convection is such that the concentrations are maximized. It seems likely that these hotspots are influential in soot filter regeneration processes. However, the frequency and location of these hotspots would appear to be quite dynamic as they are inherently coupled to the soot cake layer and the subsequent redistribution of the flow field. Can it be that these types of interplays could help explain the phenomenon reported by Opris (14) in which the balance point temperature was found to be dependent upon the soot loading level?

CASE 2

The second case explored was an extension of the first case. It was observed that the perpendicular flow velocity of the gases to the soot cake layer was not entirely uniform in the first case. This led to the postulation that soot deposition would, therefore, not be uniform across the cake layer as well. Presumably the soot deposition rate would be greatest at those locations where the perpendicular flow velocities were the greatest. This would imply that the cake layer would grow faster in these locations until the cake layer growth would force a redistribution of the flow velocities. In fact, the results indicated that the face velocities did increase in the troughs, figure 9, indicating that the cake layer is a dynamic structure. Presumably, even in a steady state engine operating condition one would find a perpetually evolving soot cake with oscillating periods of growth and consumption. In-situ testing should help to validate these hypothesis.

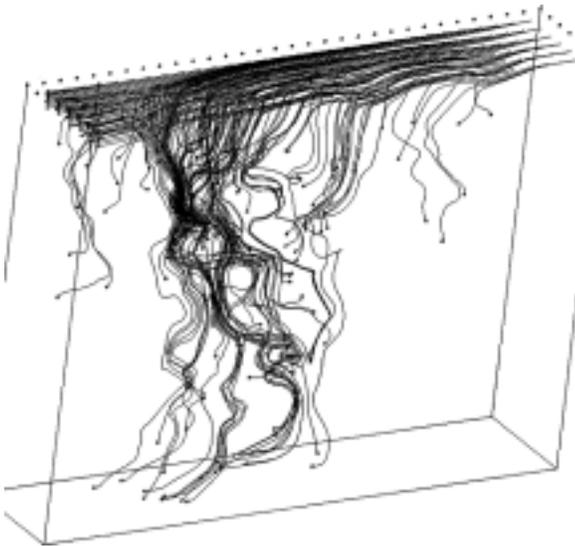


Figure 9 "streak lines – test Case 2"

SUMMARY

This paper presented the first status report of a multiyear project at PNNL focusing on the trapping and oxidation of diesel particulates. The initial focus of this program is on the subgrid modeling of filtration devices. Descriptions of the modeling technique, the initial kinetics, and the development of the computational domain were provided.

We have created a subgrid physical domain and an operable lattice Boltzmann model for a standard cordierite particulate filter. The filter chosen for our program is a commercially available design produced by Corning Inc. under the trade name EX-80. This filter substrate was chosen as it is perhaps the most common design in use today.

Our approach has been to model a section of the filter surface with dimensions on the order of 100 microns to the side. This length scale was chosen to accommodate the computational fluid dynamic codes which are used for device scale modeling. These device scale CFD models typically have a minimum length scale for the discretized elements of 100 microns. However, this length scale is far greater than many of the fundamental length scales of the physical problem, e.g. pore diameters, soot cake diameter, soot particle sizes... Therefore, these codes must make gross approximations to average these effects over the individual computational element. Our work at these smaller length scales may help elucidate phenomenon and help formulate proper approximations for the device scale CFD codes

The physical domain has been completed, and initial chemical kinetics for soot oxidation are being employed in the benchmarking stage of program execution. Example Lattice-Boltzmann computations have illustrated the potential utility of this approach. Future activities will focus on further characterization of soot deposition and cake structure phenomenon as well as in-situ model validation experiments.

FUTURE DIRECTION

- Empirical validation
 - in-situ, sub-micron scale experiments
 - device scale validation in conjunction with established CFD techniques
- Refine the representation of the cake layer structure
- Refine the representation of the kinetics
 - soot oxidation
 - gas phase chemical kinetics
- Apply technique to other aftertreatment technologies
 - fibrous or metallic soot filters
 - flow through devices (adsorbers, SCR...)

ACKNOWLEDGMENTS

We would like to thank the Pacific Northwest National Laboratory for the internal funding of this work (LDRD). We would also like to thank Professor John Johnson of the Michigan Technological University for his valuable comments and suggestions.

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