Transport and Reaction Phenomena in Diesel Particulate Filters

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Trends in Diesel Particulate Filter Technology for OEM applications

2000 DPF series (re-)introduction. Uncoated DPF. Soot oxidation assisted by ceria/iron based fuel-borne catalyst.

- 2003 Introduction of catalyst coated DPF. PGM-based catalysts aiming at NO to NO₂ conversion and NO₂-assisted oxidation of soot (indirect action of catalyst).
- 2010 Reduction/elimination of PGM. Higher porosity substrates to accommodate larger catalyst loads for multifunctional catalysts. Targeting of direct soot oxidation by oxygen transfer from base metal oxide catalysts. Better soot-catalyst contact.

The multiple scales of DPF systems



Konstandopoulos & Kostoglou (1999)

Current Focus: Wall Scale

Transport Phenomena

- Digital Materials and MicroFlow Simulation
- Reaction Phenomena
 - Direct catalytic soot oxidation
 - Wall-scale reaction models

Why is MicroFlow simulation needed?

- Effect of porous structure on flow resistance properties (bare and coated media)
- Effect of porous structure on filtration properties (bare and coated media)
- Soot distribution in the porous wall extent of contact with catalytic coating
- Effect of microstructure on soot oxidation kinetics
- Optimization of placement of multi-functional catalysts

There is need for a properly defined computational domain at the microscale: DIGITAL MATERIAL

Creation of Digital Materials

Different reconstruction "recipes" for:

- **granular** (e.g. SiC)
- reaction-born (e.g. cordierite)
- **fibrous** (e.g. metallic & ceramic fiber)
- **foam/cellular** (e.g. alumina & metal foam)
- combinations of fibrous and granular media (e.g.sintered metal)

 Validation criteria: matching different microstructural descriptors between real and digital materials

Creation of Digital Materials

Granular (SiC)





Reaction formed (e.g. cordierite)



Sintered Metal



Fibrous



Acicular



Validation: granular material





digital material



Validation: reaction-born material



Validation: foam material



digital material



Creation example: fibrous digital material

Created according to specification of fiber diameter and fiber orientation distribution



Flexible Fibers

Bonded Fibers



Creation example: sintered metal filter

Combination of fibrous and granular stuctures



cross-section



Konstandopoulos et al. SAE 2005-01-0968

Deposition in filter wall: soot concentration





Size Specific Filtration Efficiency



Application to SiC DPF characterization



New effective length scale to correlate monotonically the filtration efficiency of SiC DPF

SAE Paper 2007-01-7346



Direct Catalytic Soot Oxidation

Soot-Catalyst Contact Recognized as Major Factor since 1981

Murphy et al SAE Tech. Paper No. 810112 Re-introduced in Neeft (1995)

Fuel Borne Catalyst (FBC)





Catalyst Coated Filter



Soot oxidation on Catalyzed Filters under isothermal conditions

Oxidation is not sustained throughout the conversion!



Sustained Soot Oxidation at 530 C

Ce/Fe-based fuel borne catalyst (FBC)



SAE Paper 2007-01-7333

Sustained Soot Oxidation at 590 C

Ce/Fe-based fuel borne catalyst (FBC)



SAE Paper 2007-01-7333

Soot deposited on catalyst coated filter

Two different filters - Substrate 1 and Substrate 2

- Ceramic honeycomb wall-flow segments (34 mm x 50 mm)
- Same cell density and wall thickness
- Different pore structure

Coating method

Conventional

Advanced (promoting contact of soot and catalyst)

Soot oxidation on catalyzed filters



Soot Oxidation on Catalyzed Filters



The Two-Layer Model (1998-99)

Layer 2: Soot "layer"

Soot particles form a "queue" on top of the filled-up catalyst layer. They can enters into catalyst affected layer depending on an interaction parameter $\xi \in [0-1]$.



Filter wall

Very small amount of particles stay in the wall of catalyzed DPFs. Exponentially distributed across the wall thickness.

Layer 1: Catalyst affected "layer"

Catalyst coating can interpenetrate/overlap partially with top part of the wall. Hence better to describe it in terms of a spatial "field of catalyst activity".

Particles oxidize when found within the "field of activity" are considered *in contact* with the catalyst. Fraction $\beta \in [0-1]$ of the soot surface` in contact with the catalyst depending on coating structure.

Konstandopoulos and Kostoglou AVCS 1998 Konstandopoulos and Kostoglou SAE Tech. Paper No. 1999-01-0469

The Two-Layer Model Equations



$$\frac{dm_2}{dt} = -(k_{th}m_2 + \xi(\beta k_{cat}m_1 + (1-\beta)k_{th}m_1))$$

For FBC only Layer I is applied

Soot oxidation rate based on Two-Layer Model



Conversion: Two-Layer Model vs. Experiment



Two-Layer Model vs. Experiment



Kinetic parameters for catalyzed DPFs

No.	Sample	β	ξ
1	F6014PS	0.032	0.195
2	F6007PV	0.075	0
3	F6012PV	0.064	0.642
4	F8012A	0.008	0.133
5	F8015P1	0.024	0.133
6	F80M6U	0.0014	0.127
7	F8016W	0.147	0.290
8	F8018A	0.019	0.343
9	F100M4A	0.017	0.213
10	Μ	0.214	0.013
11	Т	0.0006	0.013

Modeling of Multifunctional Wall-Flow Monoliths: Kinetics Splitting Scheme



SAE Paper 2007-01-1130

Kinetics Splitting Scheme Features

- Exploit the large difference in reaction kinetics among the species reacting with oxygen to order the reactions in "sublayers"
- Assume first order kinetics to derive the oxygen consumption in each reaction path taken to occur sequentially
- Result is globally analytical but approximate expressions that provide the overall species concentrations across the filter wall
- This approximation is validated by comparison to the exact numerical solution
- It can be embedded in multidimensional simulators to achieve computations of multifunctionally catalyzed DPFs, with 3-D spatial patterning of the catalysts at a fraction of the time needed with existing numerical formulations
- It makes possible, due to its analytical structure, the deployment of efficient on-board monitoring and control algorithms, implementable in computationally limited engine control units (ECUs).

Validation of Kinetics Splitting Scheme HC and CO conversion vs Temperature

Case 1: 10,000ppm HC/ 2,000 ppm CO/ 10% O₂

Case 2: 2,000 ppm HC/ 2,000 ppm CO/ 10% O₂

Case 3: 2,000 ppm HC/ 2,000 ppm CO/ 2% O₂

Numerical vs. Analytical Approximation



Validation of Kinetics Splitting Scheme Reaction Rates of soot oxidation by NO₂

Numerical solution and Kinetics splitting scheme approximation are indistinguishable



SAE Paper 2007-01-1130

Conclusions

- Digital materials methods are important tools for fundamental studies of DPF microstructures and to guide future material development.
- The study of conversion dependent phenomena during isothermal catalytic soot oxidation reveals that geometry is more important than chemistry for sustained soot oxidation in catalytically coated filters.
- The influence of soot-catalyst proximity, on catalytic soot oxidation in catalyzed prototype filters is found to be excellently described (+/- 5%) by the so-called Two-Layer model, developed previously by the authors.
- An efficient analytic wall-scale reaction model based on a kinetics splitting scheme was derived and validated against numerical integrations of the coupled species equations.
- The development of direct soot oxidation catalytic filters requires close integration of porous substrate and catalytic coating technologies in order to optimize soot-catalyst contact, including the dynamics of its evolution.

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Dedication

to the loving memory of Professor Sheldon K. Friedlander



APT Lab 10 year anniversary Thessaloniki May 20, 2006