Combination and Integration of DPF – SCR Aftertreatment Technologies

P.I. – Kenneth G. Rappé
Darrell R. Herling
John Lee, John Frye, Gary Maupin
Pacific Northwest National Laboratory (PNNL)

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ACE025

Pacific Northwest
NATIONAL LABORATORY

OVERVIEW

Timeline

- Start – Oct 2008
- Finish – Oct 2012
- 37% complete

Barriers

- Barriers addressed
  - Heavy truck thermal efficiency
  - Aftertreatment cost
  - Combined NOx and PM emissions

Budget

- Total project funding
  - $1.6M DOE share
  - $1.6M I.K. Contractor contr.
- $200K received in FY09
- $400K received in FY10

Partners

- Primary Partner: PACCAR
  - PACCAR Technical Center
- DAF Trucks (operating as an extension of PACCAR)
  - Utrecht Univ. operating as a supportive entity to DAF
- Project Lead: PNNL

OBJECTIVES

Fundamentally understand the integration of SCR & DPF technologies to provide a pathway to the next generation of emissions control systems

- Probe interaction of DPF-SCR couples to better understand the optimization of the coupled units
- Determine system limitations, define basic requirements for efficient on-board packaging and integration with engine
- Develop an understand of …
  - optimal loading of SCR catalyst for maximizing NOx reduction while maintaining acceptable ΔP and filtration performance.
  - proper thermal management of the system for regenerating the DPF without negative impacts on the SCR catalyst.
  - SCR aging, including effect of …
    - locally higher temperatures of soot combustion.
    - active site blockage.
    - zeolite structure integrity.
    - metal migration.

MILESTONES

- Identify approach to system integration, metrics by which success will be gauged (4 mo.) – complete
- Develop technique for integration of SCR active phase into wall-flow configuration – complete
- Demonstrate integrated DPF/SCR on 2 cm dia. wall-flow filter with synthetic diesel exhaust stream (15 mo.) – challenges with SCR active phase
- Demonstrate integrated DPF/SCR on 2 cm dia. elevated porosity filter (19 mo.) – challenges with SCR active phase
- Prepare integrated DPF/SCR on 15 cm dia. filter (30 mo.)
- Discussions with manufacturer on pathway to fabricate integrated DPF/SCR for vehicle demonstration (33 mo.)
- Demonstrate integrated DPF/SCR on 15 cm dia. wall-flow filter on diesel engine slip stream (39 mo.)

APPROACH/STRATEGY

DOE Merit Review
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- Flow restriction concerns
  - ∆P: SCR/DPF > SCR + cDPF
  - Back pressure dependant on filter type & washcoat loading
  - Focus on different filter substrates & SCR washcoat loadings to maximize NOx reduction performance & minimize flow restriction
- Optimal SCR catalyst loading
  - Versus effect on permeability and DPF filtration performance
- Thermal management
  - Minimizing impact on SCR catalyst
- Evaluation SCR catalyst impact via detailed system interrogations (Utrecht)
- Address NOx conversion with accumulated soot
  - Active site blockage, soot-combustion facilitated thermal aging, etc.

TECHNICAL ACCOMPLISHMENTS

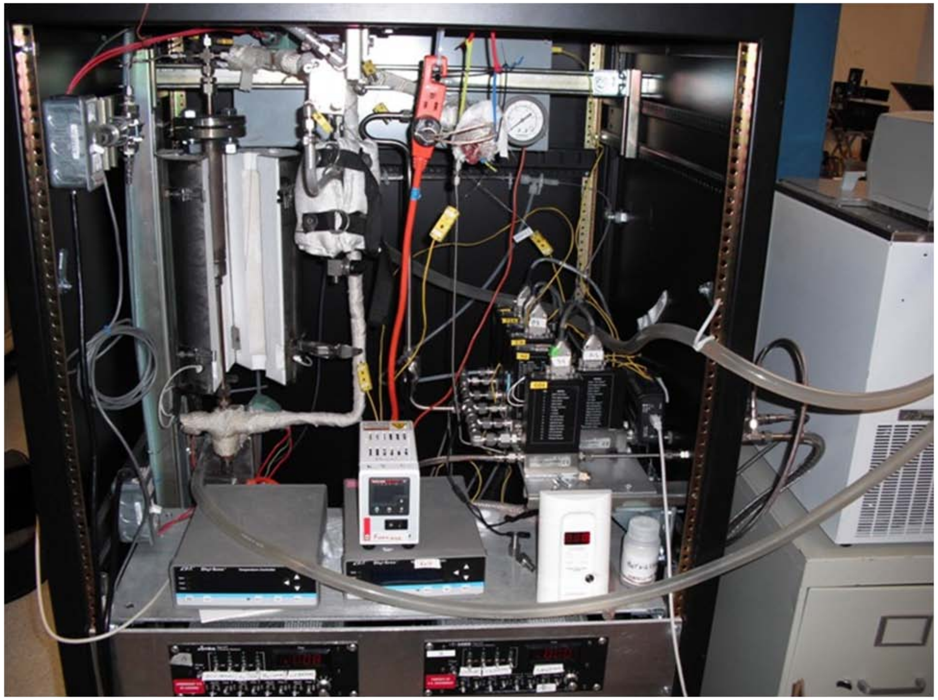
Detailed filter substrate evaluation

  - Cordierite, SiC, Al_2TiO_5, ACM, SiN

- Key attributes for integrated system
  - Pore characteristics: open, uniform structure w/ good mech. strength
  - Thermal conductivity, heat capacity
- Cordierite – primary
  - Fast warm-up
  - Lower melting point
  - Less controlled pore structure
- Silicon Carbide (SiC) – secondary
  - Increased cost (low TSP mitigated by segmentation)
  - Higher heat capacity (higher soot loading, longer warm-up)
  - Higher thermal conductivity (better SCR protection?)
  - Favorable uniform & open pore network

TECHNICAL ACCOMPLISHMENTS

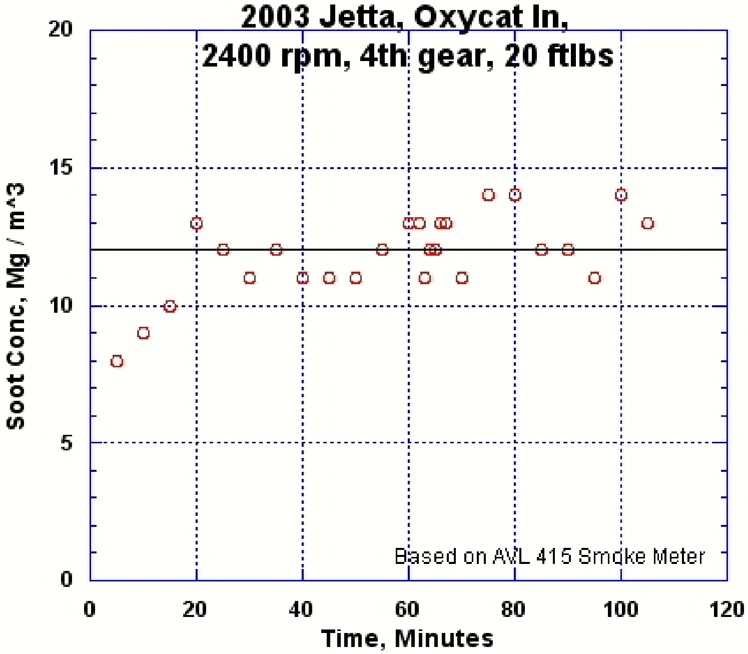
- Sample core testing capability
  - For evaluating sample core performance
    - integrated system performance
    - activity – regeneration – activity
  - Regeneration strategy development/evaluation



TECHNICAL ACCOMPLISHMENTS

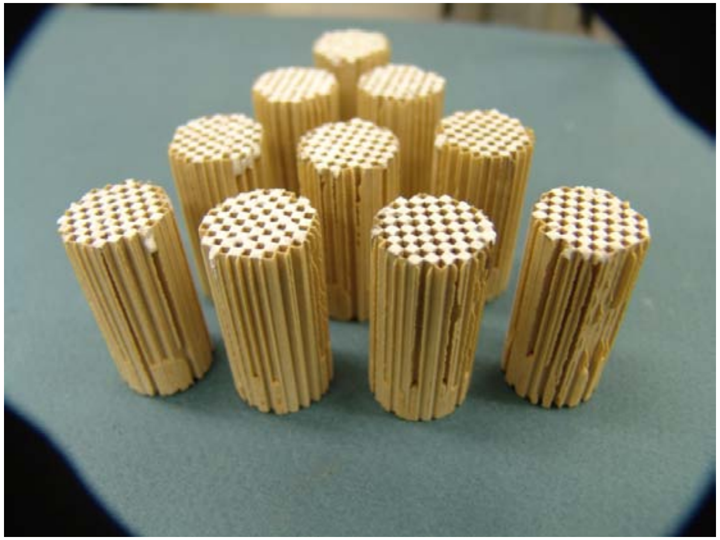
Sample Soot Loading

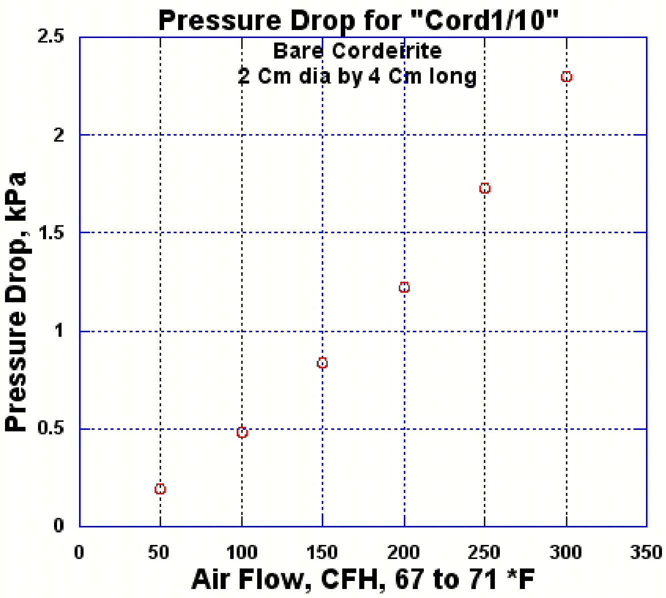
- Loading filters with 2003 VW Jetta exhaust
- Loading based on AVL 415 Smoke Meter measurements
  - Targeting engine condition producing ~12 mg/m^3
  - Good reproducibility



Sample Core Preparation

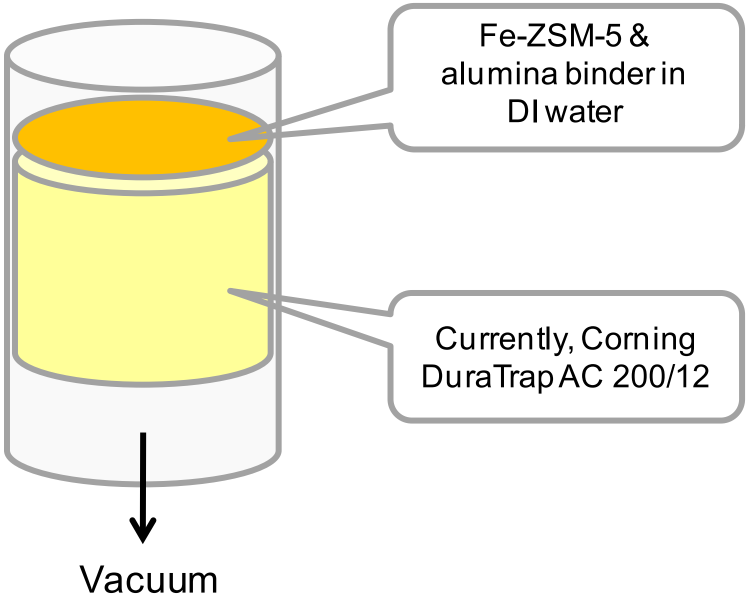
- Sample cores prepared for coating & lab reactor testing
- Coating efforts guided by back-pressure measurements
  - Washcoat and measure ΔP increase

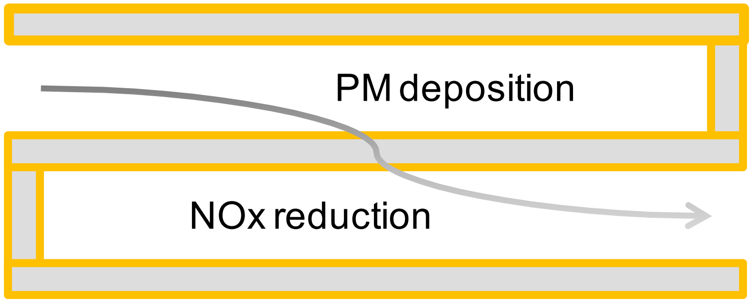




Washcoating Plan

- Sample preparation method development
  - Highly iterative coating process (pull-blow-dry-measure)
- Vacuum – suction method

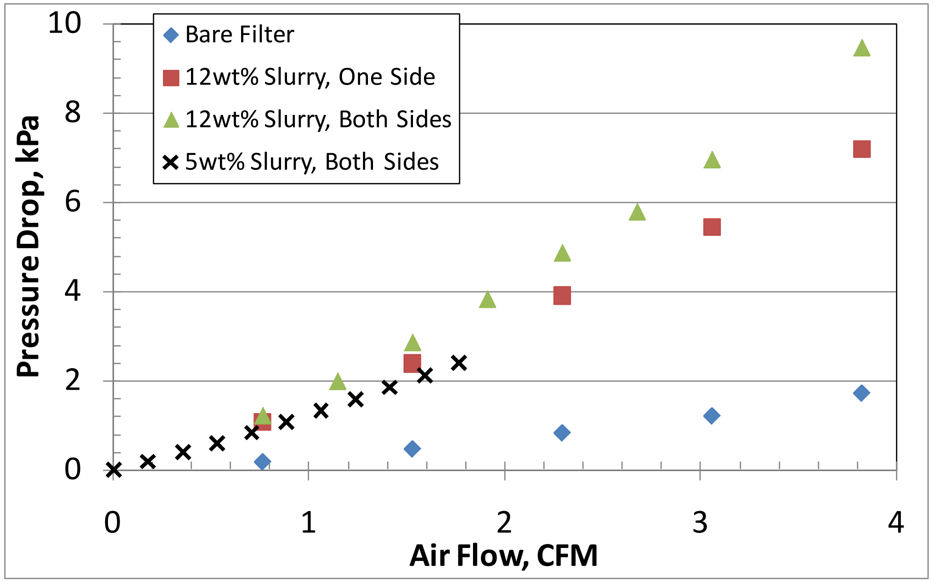




- Washcoat on outlet or both channels of various filter media
- Compare ΔP, NOx reduction & “passive” soot oxidation performance

TECHNICAL ACCOMPLISHMENTS

Filter Core Coating – blank zeolite



- 12 wt % solids slurry
  - Coating one side – 5.73 wt% loading – 32 g/L catalyst incorporated
  - Coating both sides – 10.24 wt% loading – 60 g/L catalyst incorporated
- 5 wt % solids slurry
  - Coating both sides – 5.51 wt% loading – 31 g/L catalyst incorporated
- **Method adjustment → improved filter permeability**

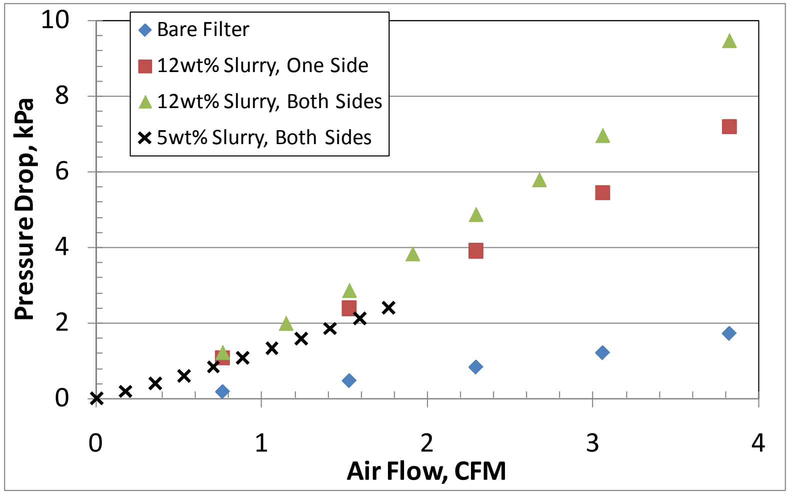
Filter Pressure Drop Scaling

- $\Delta P = \Delta P_{\text{filter wall}} + \Delta P_{\text{soot layer}} + \Delta P_{\text{inlet/outlet channel}} + \Delta P_{\text{entrance/exit}}$
  - Clean filter: $\Delta P_{\text{soot layer}} = 0$
  - $\Delta P_{\text{entrance/exit}}$ typically $O(10^{-2}\text{-}10^{-3})$; can be neglected with minimal consequence
  - $\Delta P_{\text{inlet/outlet}}$ a function of filter characteristics and exhaust gas conditions; unaffected by filter wall conditions
  - $\Delta P_{\text{filter wall}}$ a function of filter wall permeability, k_0

Approach to scaling pressure drop

- Catalyst wash coat → decreased permeability (k_0) through filter wall
- Full-size filter pressure drop – predicted via quantitatively determining the effect of the catalyst wash coat on the filter wall permeability

Filter Pressure Drop Scaling – 900 SCFM, 450°C

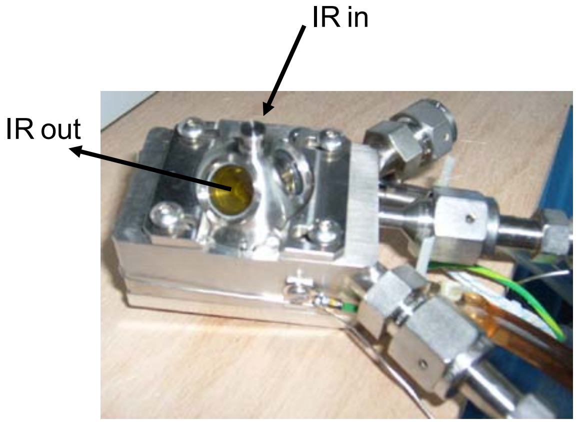


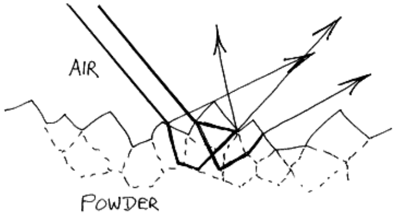
- filter wall permeability, k_0 (200 cpsi, 12 mil wall)
  - ~$5.3\text{x}10^{-13}\ m^2$ for a typical fresh cordierite filter (~48% porosity)
    - Filter wall – ΔP = 1.13 kPa
    - Inlet/outlet channel effects – ΔP = 4.17 kPa
  - ~$1.56\text{x}10^{-13}\ m^2$ for 12wt% slurry coating (60 g/L catalyst loading)
    - Filter wall – ΔP = 3.83 kPa
    - Inlet/outlet channel effects – ΔP = 4.17 kPa

COLLABORATIONS

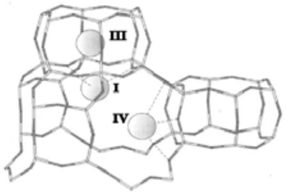
University of Utrecht

- In-situ examinations, active site analysis, system aging analysis





- In situ cell from Harrick
- Powder → easy sample preparation
  → similar conditions as in activity studies
- Surface species under reaction conditions
- Product gas analysis by GC and MS



- Use high Silica zeolites
- Locate active sites in zeolite
- Powder studies
- Co-relate structural features (pore/window size) to activity, eg. comparison of ZSM-5 with zeolite Y

In situ powder studies

Combined XRD/XAS/Raman

Cryostat cooling

(regular SCR ramps on powder samples with instant quenching from 400 ^{o}C to -200 ^{o}C)

Monolith Imaging (synchrotron)

μ-diffraction/fluorescence

↓ 2D phase mapping along x and y

↓ 2D elemental mapping along x and y

↓ Rietveld analysis
- cation migration
- framework geometric distortion
- MO species
- strain due to ageing

- Samples: 1 mm x 4mm (cut sections)
- Aging: Different conditions (0-250 h)
- 5 – 10 μm step sizes (x,y)
- Experiments done at RT
- **Coated monolith from PNNL**

FUTURE WORK (short term)

- Method development with SCR active phase
  - Employing 200 cpsi, 12 mil wall cordierite DPF
  - Efforts guided by ΔP measurements, sample mass increase
  - Testing of samples to include
    - SCR performance (fresh)
    - DP, activity versus soot loading
    - Regeneration investigations (active & passive)
    - Hydrothermal aging, SCR performance
- Method development with elevated porosity DPF
- **SCR active phase**
  - Preference is to use vendor-supplied commercial catalyst
  - To date, significant challenges in acquisition has slowed program progress substantially
  - Currently attempting to move forward with internal formulation

SUMMARY

- Method developed for sample preparation of coupled SCR – DPF systems
- **Key parameter is maximizing SCR active phase loading with acceptable filter wall permeability**
- Tools are in place for detailed interrogation of active coupled systems, including:
  - SCR performance
  - Soot loading
  - Regeneration investigations
- Method and proper metrics will allow scale-up of coupled systems for ultimate engine demonstration