Combination and Integration of DPF – SCR Aftertreatment Technologies

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ACE025

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OVERVIEW

Timeline

- Start – Oct 2008
- Finish – Oct 2012
- 58% complete

Barriers

- Barriers addressed
  - Cost effective emission control
  - Heavy truck thermal efficiency
  - Combined NOx and PM emissions

Budget

- Total project funding
  - $1.6M DOE share
  - $1.6M I.K. Contractor share
- $200K received in FY09
- $400K received in FY10
- $275K received in FY11

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.) – complete
- Demonstrate integrated DPF/SCR on 2 cm dia. elevated porosity filter (19 mo.) – complete
- Prepare integrated DPF/SCR on 15 cm dia. filter (30 mo.) – discussions underway with BASF on value of this step versus going straight to full-size prototype.
- Discussions with manufacturer on pathway to fabricate integrated DPF/SCR for vehicle demonstration (33 mo.) – discussions underway
- Demonstrate integrated DPF/SCR on 15 cm dia. wall-flow filter on diesel engine slip stream (39 mo.)

APPROACH/STRATEGY

- Flow restriction concerns
  - ΔP: SCR/DPF > SCR + cDPF
  - Back pressure dependant on filter type, filter specifications, washcoat technique & ***loading***
  - Maximize NOx reduction performance, maximize PM filtration performance, minimize flow restriction
- Optimal SCR catalyst loading
  - Versus effect on filter permeability, particulate filtration performance, DPF regeneration performance
- Thermal management
  - Minimizing impact on SCR catalyst
- Detailed interrogations evaluating SCR catalyst impact (Utrecht)
- Address NO_x conversion with accumulated soot

APPROACH/STRATEGY

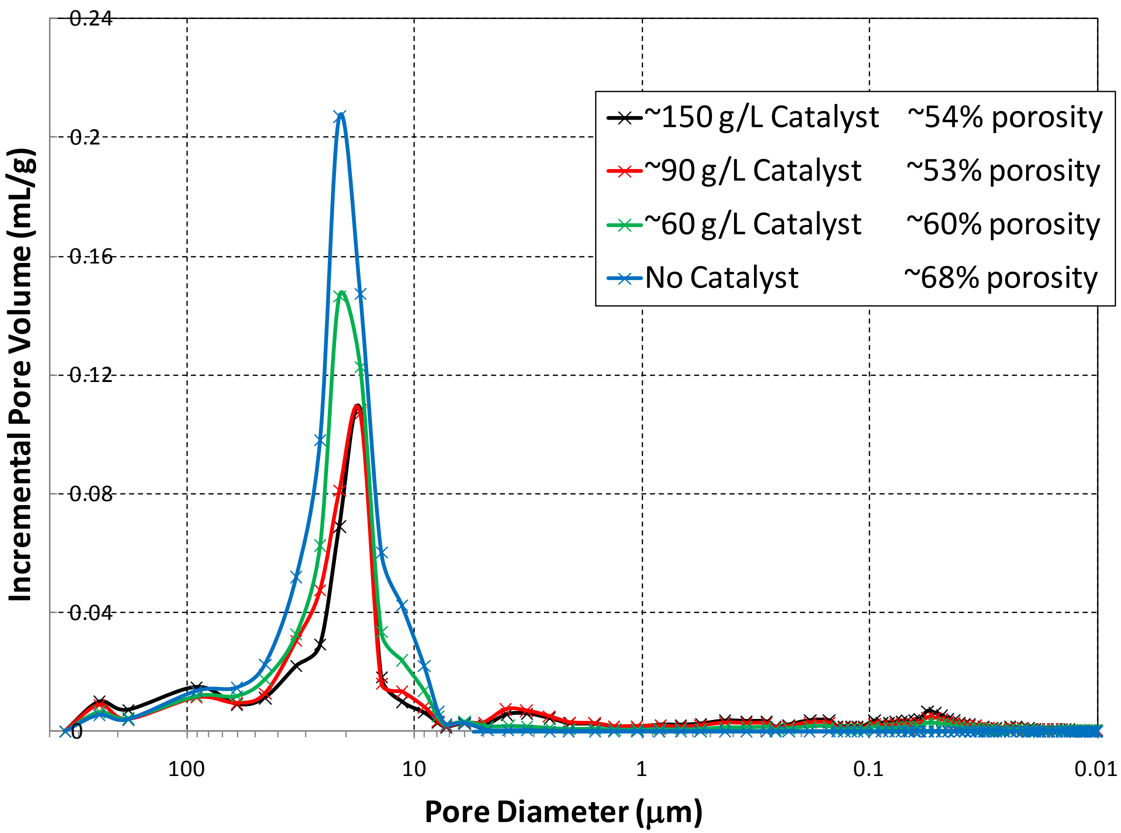
- First key barrier to overcome for system implementation: *back pressure*
  - Solutions: high porosity substrate, refined wash-coating technique
    1. Higher porosity substrate
    2. Refined wash-coating technique

1. Vendor-supplied ultra-high porosity (UHP) substrate
   - NDA put in place with Vendor; developmental substrate
   - UHP filters acquired by PNNL for integration effort

2. Supplier wash coating technology
   - NDA in place with Catalyst Supplier
   - Supplier SCR catalyst technology
   - ***Coated parts in hand, testing and materials analyses underway***
   - ***Developing excellent working relationship with Vendor's Heavy Duty Systems R&D group***

Physical examinations of integrated system

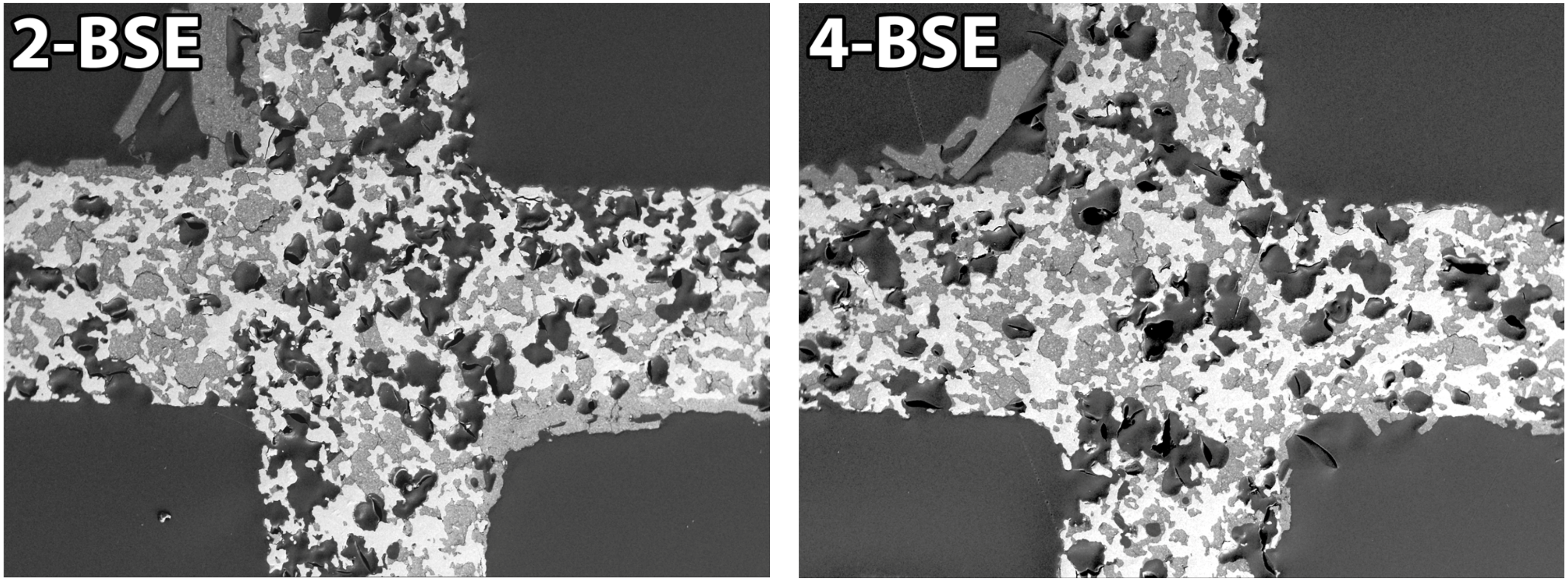
▶ Pore characteristics – Hg porosimetry



- Catalyst appears to *fill* (or plug) 8 – 50 mm pores, versus *coat* pores.
- If catalyst was largely present as a coating, we should see a more significant shift of the curve asymmetrically to smaller pore sizes.
- The majority of >90 g/L does not appear go into the pore structure, only goes into a *small number* of very large pores (>20 mm).

Physical examinations of integrated system

- Washcoat interrogation – SEM imaging ~150 g/L loading



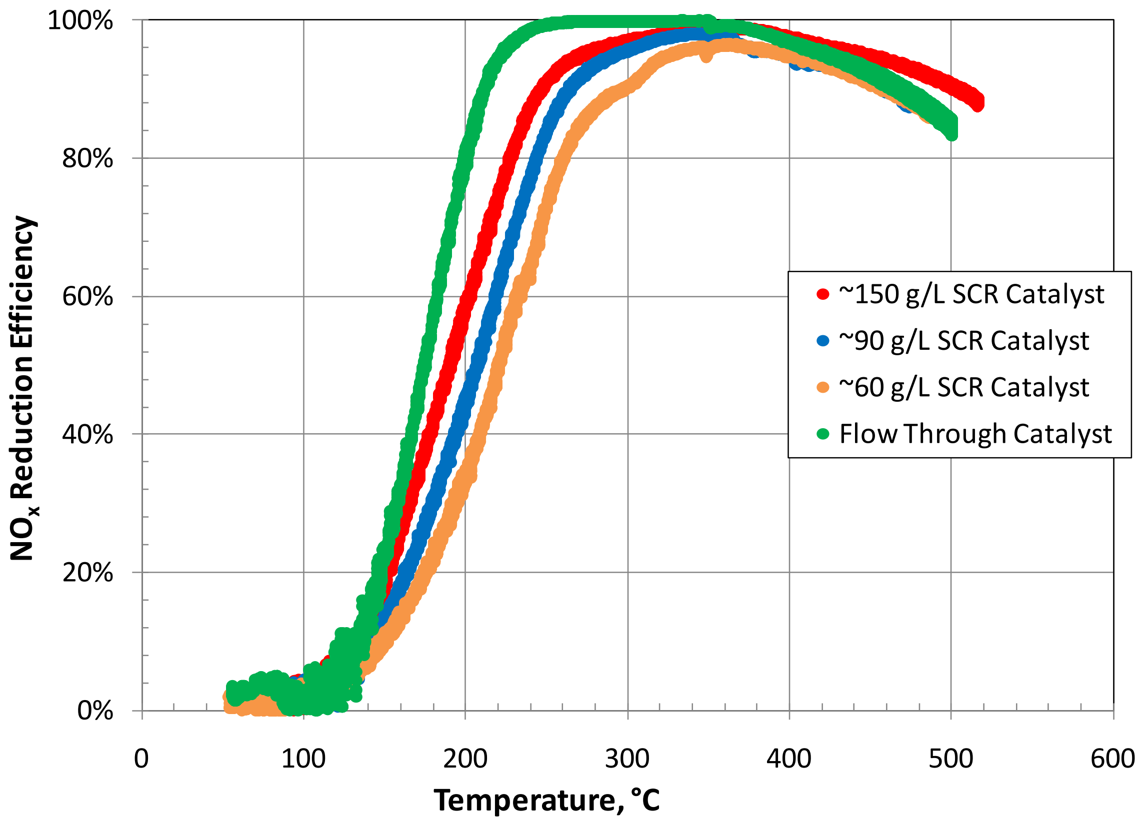
- Confirms Hg porosimetry results
  - Where catalyst exists, filling pores versus coating pores.
  - At ~150 g/L, significant catalyst visually evident on channel wall
- Continue imaging f()=catalyst loading, refined technique

TECHNICAL ACCOMPLISHMENTS

SCR reaction investigations

1. **Effect of configuration (& catalyst loading) w/o soot**

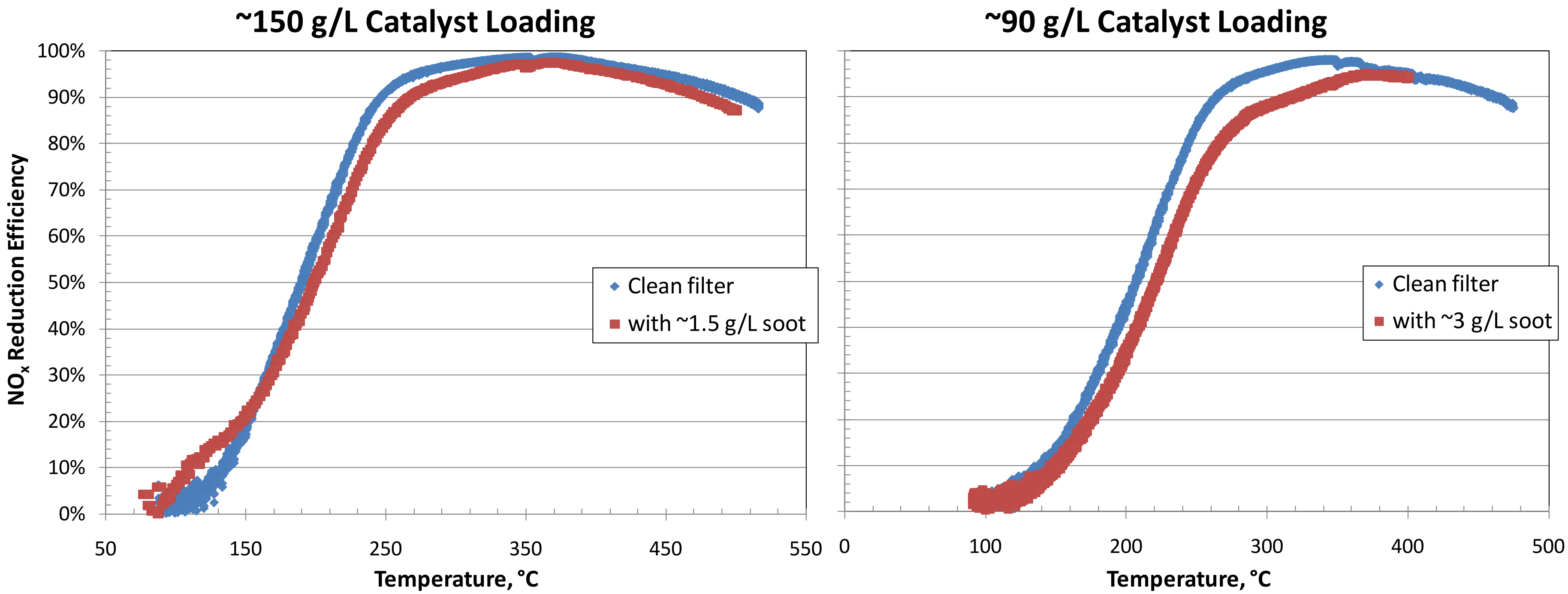
500 ppm NO & NH_3, 35K GHSV



- Appears to be an effect of configuration (i.e. flow through versus wall flow)
- Increased activity of 150 g/L versus 90 g/L suggests catalyst present on filter channel walls has benefiting effect.
- Suggests refined wash coating technique with SCR catalyst within wall, on outlet channels, etc., would improve activity.

TECHNICAL ACCOMPLISHMENTS

- SCR reaction investigations
  2. **Effect of the presence of soot**: 500 ppm NO & NH_3, 35K GHSV



- Effect of the presence of soot on the SCR reaction(s) is definite, but not significant

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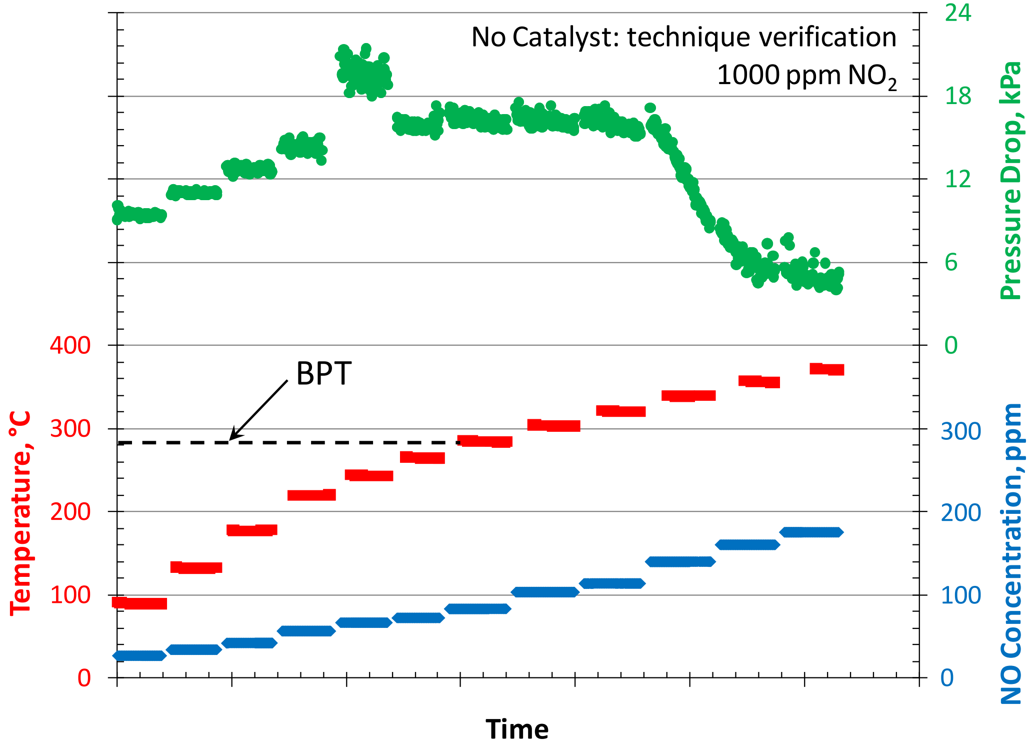
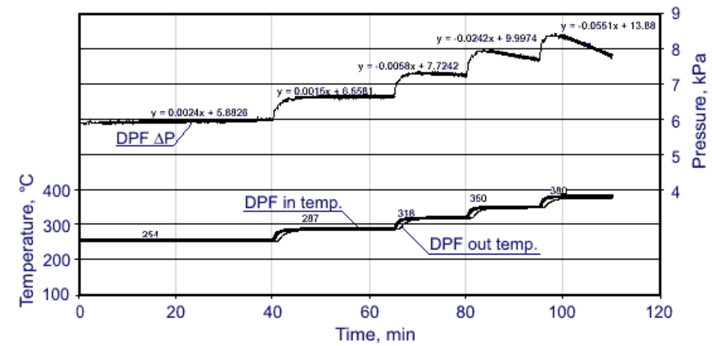
Effect of the presence of SCR reaction(s) on the passive soot reaction(s): balance point temperature (BPT) identification





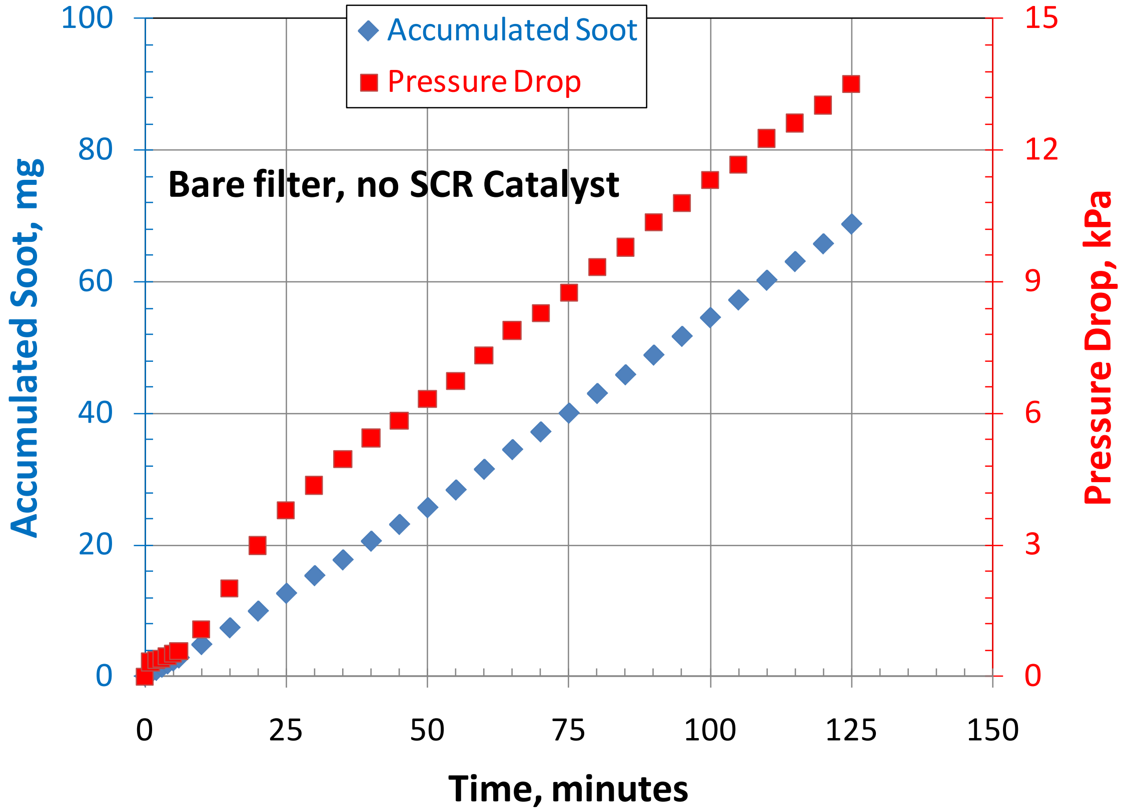
DECSE 5-mode balance point test

(*DECSE, 1999. "Diesel Emission Control Sulfur Effects Program, Phase I Interim Report No. 1", U.S. DOE, August 1999)*

- Continue BPT examination at different catalyst loadings
- BPT in the absence (no NH_3) and presence (w/ NH_3) of SCR reaction

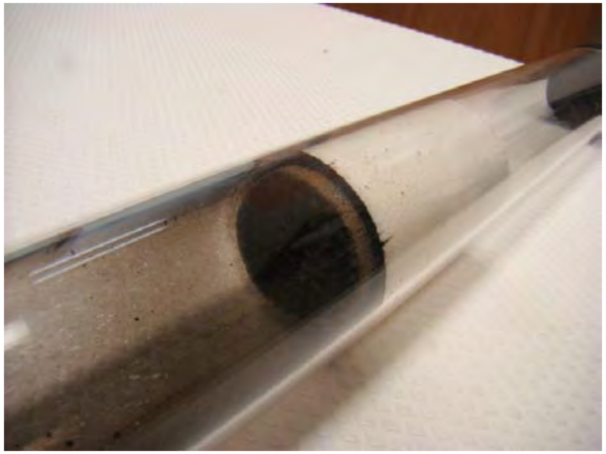
Soot loading characteristics of integrated system

- Dynamic pressure drop of filter during soot collection



Characteristics appear typical* of very high porosity filter

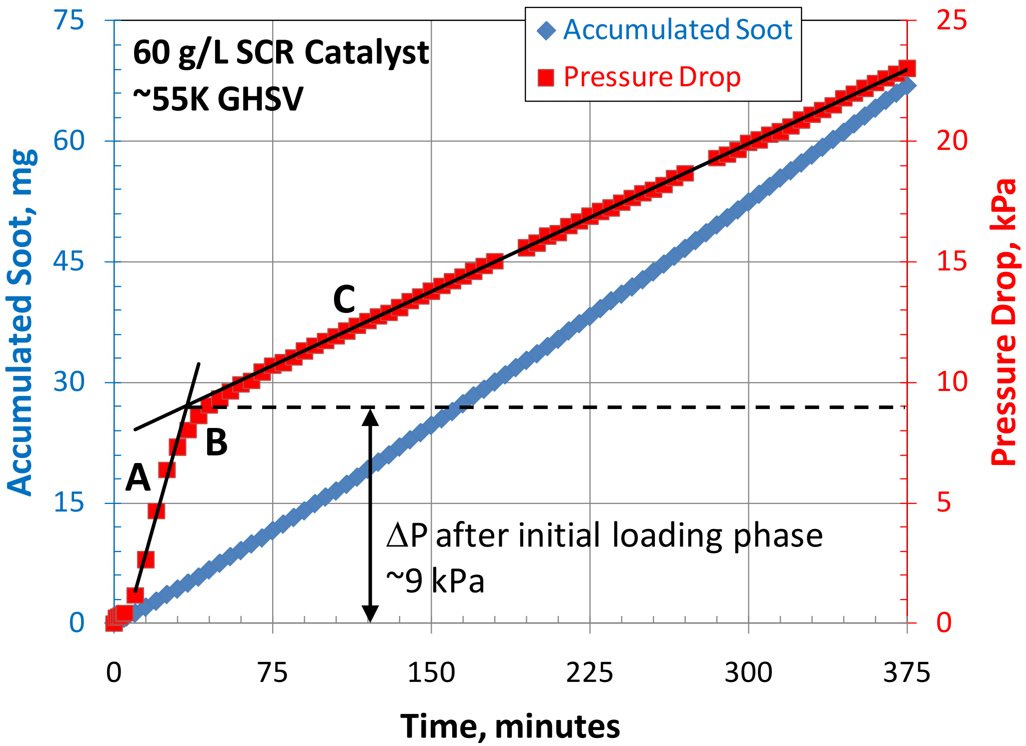
*Merkel et al, DEER 2003



TECHNICAL ACCOMPLISHMENTS

Soot loading characteristics of integrated system

- Dynamic pressure drop of filter during soot collection

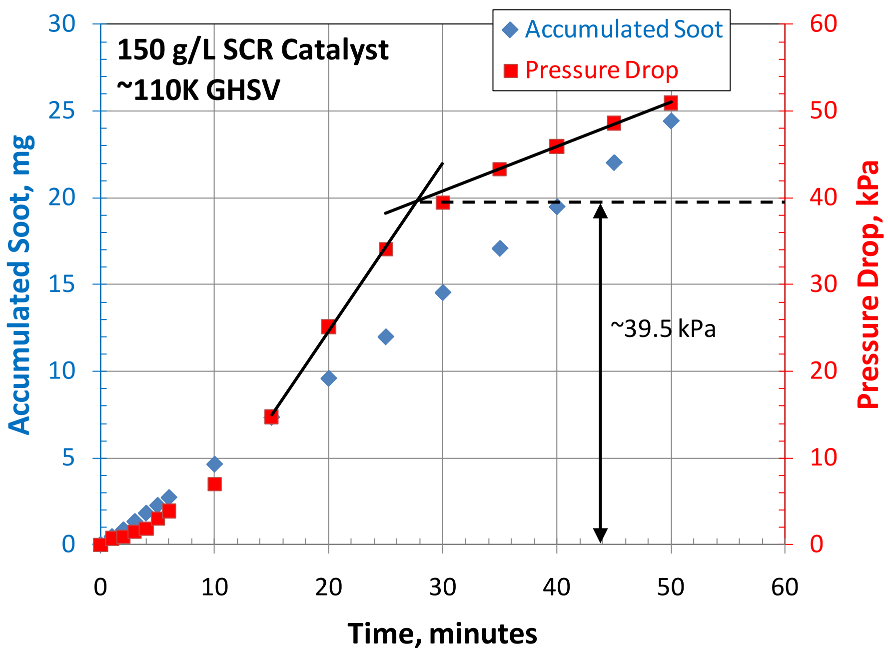


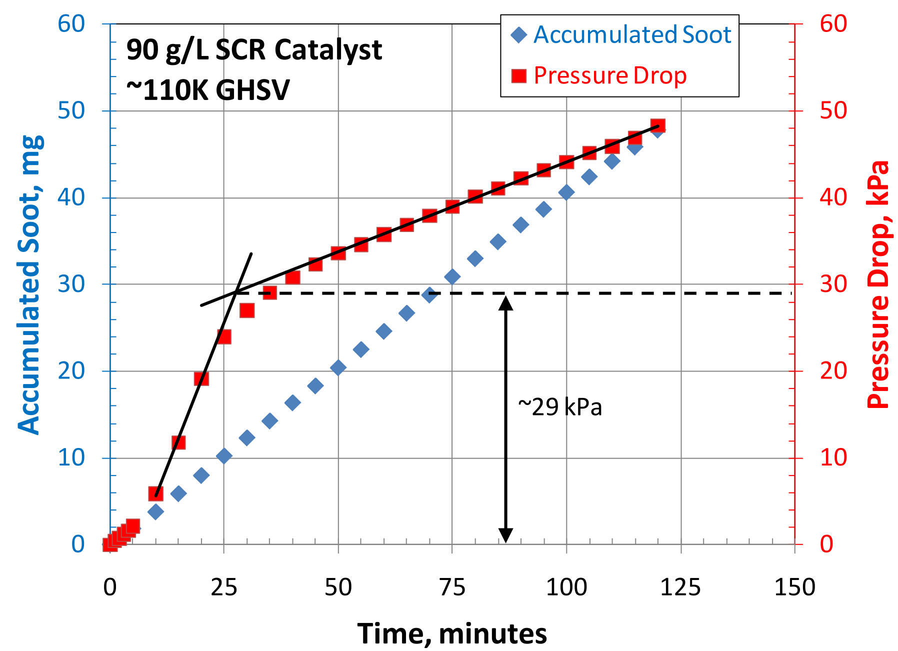
- ΔP_{clean}
  - SCR catalyst effect
- ΔP after initial loading phase
  - Dominated by depth filtration
  - SCR catalyst effect
- Loaded trap (C)
  - Largely unaffected by filter characteristics

- Good collection performance of 60 g/L integrated sample
- Performance very comparable to conventional porosity filter

Soot loading characteristics of integrated system

- Dynamic pressure drop of filter during soot collection





- As expected, increased SCR catalyst creates increased back pressure with collected soot.
- Pressure drop numbers used for predicting full-size filter performance

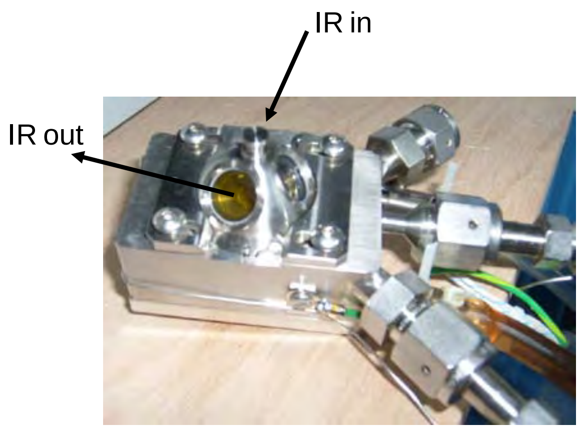
Approach to Filter Pressure Drop Scaling

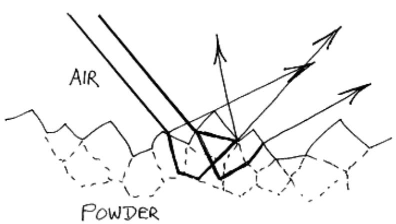
- Filter wall permeability (k_0)
- *Full-size filter pressure drop* – predicted via quantitatively determining the effect of the catalyst wash coat and dynamic soot loading characteristics on the filter wall permeability
  - Conventional (~48% porosity) filter permeability *~5.3x10^{-13} m^2*
  - Bare UHP measured filter permeability extremely high, as expected, *~18.5x10^{-13} m^2*
  - With ~120 g/L SCR catalyst, permeability still quite high *~8.4x10^{-13} m^2*
- With ~4 g/L loaded soot, filter permeability drops significantly, contributions from both filtration mechanisms (depth & cake)
  - With ~120 g/L SCR catalyst = *~0.074x10^{-13} m^2*
    - ~43 kPa predicted for full-size filter at 450 SCFM, 600K
  - With ~60 g/L SCR catalyst = *~0.15x10^{-13} m^2*
    - ~21 kPa predicted for full-size filter at 450 SCFM, 600K
- Pressure drop at ~120 g/L SCR too high. Continuing wash-coat development with Supplier

COLLABORATIONS

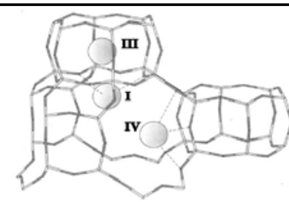
University of Utrecht

- 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 °C to -200 °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 (2011)

- Continuing wash-coat development strategy with Vendor:
  - Uniform dispersion throughout the wall
  - Loading heavy to the inlet & outlet sides and channel walls
  - Other parameters Supplier will guide on, including:
    - Varying rheological and/or wicking characteristics
    - Immersion strategy (time, repetition, etc.)
- Continuing system kinetic and performance investigations
  - Effect of soot & soot oxidation on NO_x SCR reaction
  - Effect of SCR reaction on soot oxidation
  - Integrated system soot filtration performance
  - … as a function of catalyst mass loading, catalyst configuration within filter, etc.

SUMMARY

- NDAs in place with Vendors for UHP substrate and substrate:catalyst integrated systems
- Integrated system samples in hand, currently under investigation
- Examinations interrogating the physical system as well as kinetic and dynamic performance of integrated system
  - NOx SCR performance looks good. Continuing SCR integration strategy to maximize SCR performance
  - Pressure drop performance of integrated system during dynamic soot collection is limiting. Continuing development with Supplier.
- Results of examinations will provide feedback to Supplier to guide on system integration efforts to ultimately direct towards an optimum integrated device

TECHNICAL BACK-UP SLIDES

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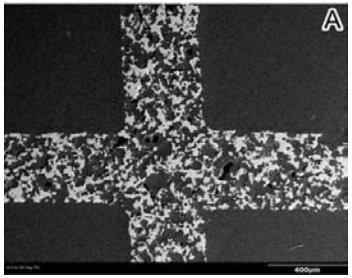
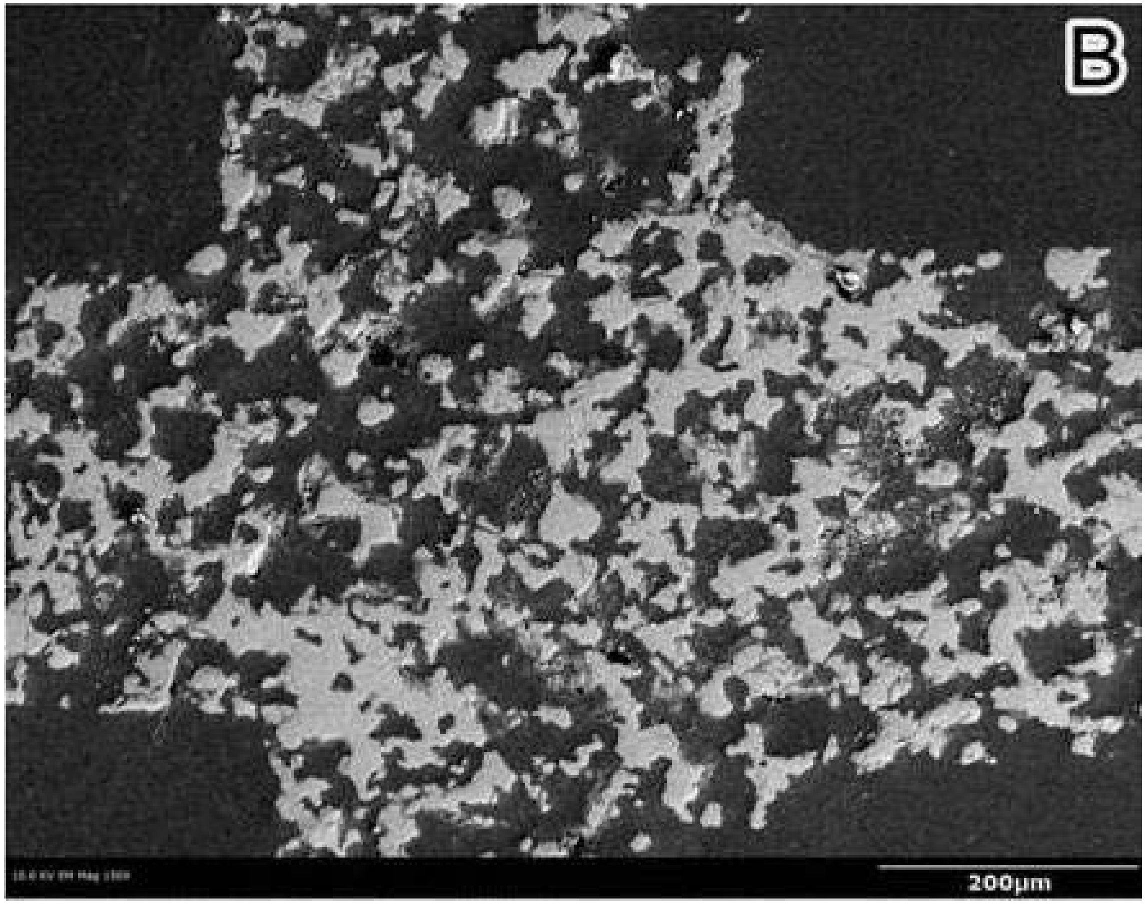
Physical examinations of integrated system

- Washcoat interrogation – SEM imaging bare filter





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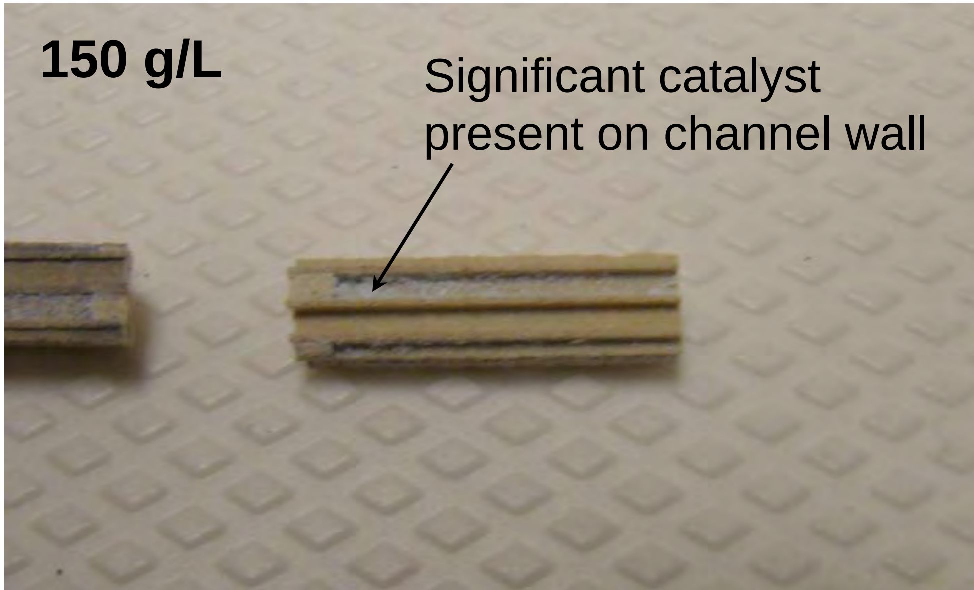
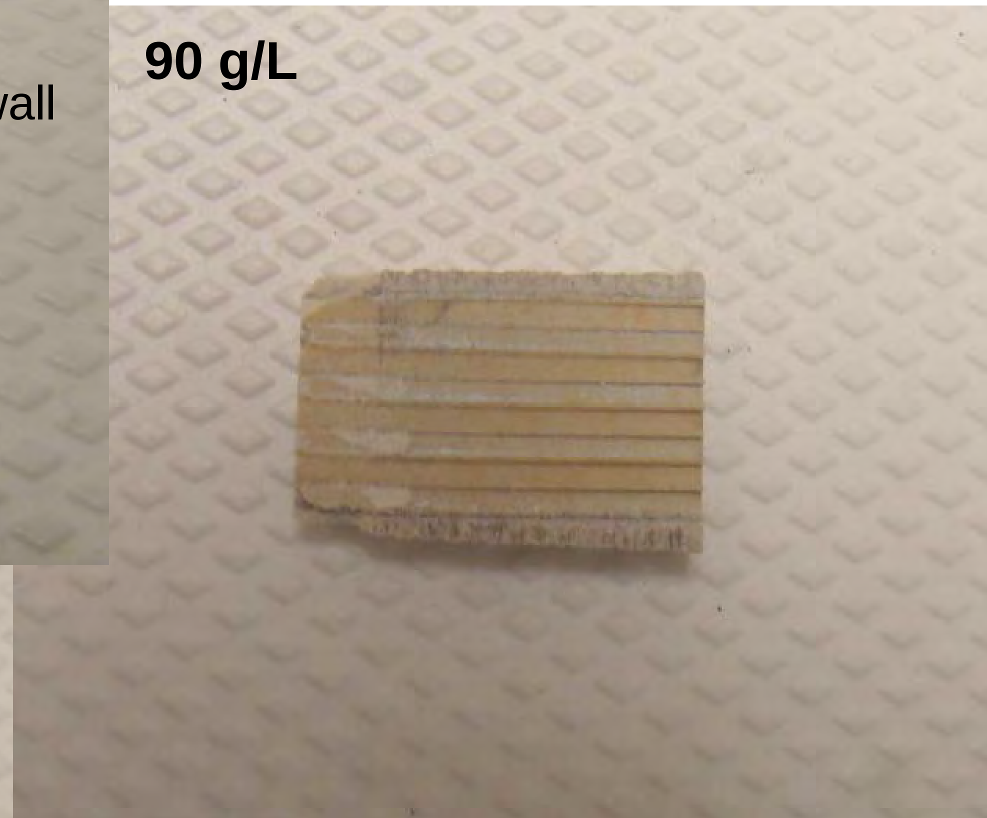
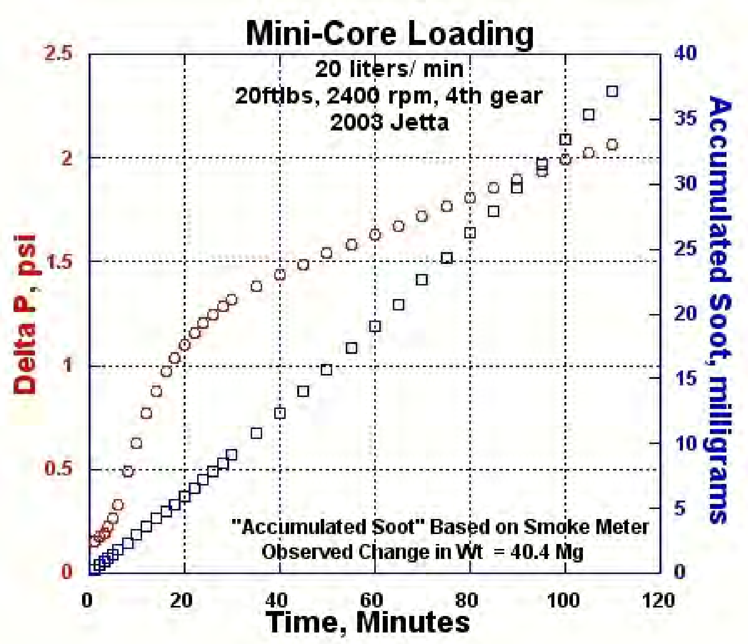
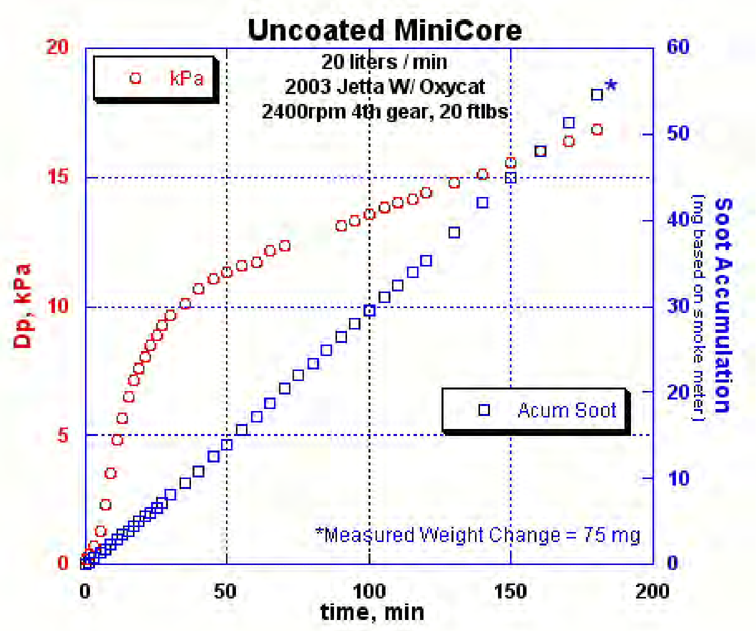




Soot loading characteristics of integrated system

- Dynamic pressure drop of filter during soot collection
  - Soot accumulation from smoke meter measurement
  - Good agreement (<10% delta) with weight change





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TECHNICAL BACK-UP SLIDES

- $\Delta P = \Delta P_{\text{filter wall}} + \Delta P_{\text{soot layer}} + \Delta P_{\text{inlet/outlet channel}} + \Delta P_{\text{entrance/exit}}$
  - Clean filter: $\Delta P_{\text{soot layer}} = 0$

$$\Delta P_{filter\ wall} = \frac{\mu_{gas} Q}{2V_{trap} N}\left[\frac{w_{wall}}{k_0 d_h}\right] \qquad \Delta P_{inlet/outlet} = \frac{\mu_{gas} Q}{2V_{trap} N}\left[\frac{8FL^2}{3d_h^4}\right] \qquad \Delta P_{entrance/exit} = \frac{\zeta_E \rho_{gas}}{2}\left[\frac{2Q}{A_{trap} N d_h^2}\right]^2$$

Q, ρ_{gas}, μ_{gas}	exhaust gas flowrate, density and viscosity, respectively
L, A_{trap}, V_{trap}	filter trap length, area and volume, respectively
N, w_{wall}, d_h	filter cell density, wall thickness and hydraulic radius (cell size), respectively
F	inlet/outlet channel friction factor (~ 28.454)
k_0	filter wall permeability ($\sim 5.3\text{x}10^{-13}\ m^2$)
ζ_E	area blockage factor ($0.2 - 0.8$), a function of d_h & w_{wall} (OFA)

$\Delta P_{entrance/exit}$	typically $O(10^{-2}\text{-}10^{-3})$ and can be neglected with minimal consequence

A. Konstandopoulos and J. H. Johnson, SAE 890405, 1989

TECHNICAL BACK-UP SLIDES

$$\Delta P_{filter\ wall} = \frac{\mu_{gas} Q}{2 V_{trap} N}\left[\frac{w_{wall}}{\boldsymbol{k_0} d_h}\right] \qquad \Delta P_{inlet/outlet} = \frac{\mu Q}{2 V_{trap} N}\left[\frac{8 F L^2}{3 d_h^{\ 4}}\right]$$

- $\Delta P_{inlet/outlet}$ a function of filter characteristics and exhaust gas conditions; unaffected by filter wall conditions
  - Assuming wash coat has no (or negligible) effect on:
    - friction factor, F
    - hydraulic radius, d_h
- $\Delta P_{filter\ wall}$ a function of filter wall permeability, $\boldsymbol{k_0}$
  - $\sim 5.3\text{x}10^{-13}\ m^2$ for a typical fresh cordierite filter (~48% porosity?)
- Catalyst wash coat → decreased permeability through filter wall
- **Full-size filter pressure drop – predicted via quantitatively determining the effect of the catalyst wash coat on the filter wall permeability**