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Development of Advanced Diesel Particulate Filtration (DPF) Systems

(ANL/Corning/Caterpillar CRADA)

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PI: ***Kyeong Lee***

Postdocs: Seungmok Choi, Heeje Seong, Hoon Lee

Argonne National Laboratory

DOE Project Managers:

Kenneth Howden & Gurpreet Singh

Office of Vehicle Technologies

Project ID: ACE024

This presentation does not contain any proprietary or confidential information

Overview

Timeline

- Start: Oct 2006
- Finish: Sept 2011 (completed)
- 95% Finished

(numerical modeling & filter structural analyses performed as extra)

Budget

- Total Project funding (5 years)
  - DOE: $2,200K
  - Industry sponsors: $1,650K
- Funding received in FY11
  - $250K

Barriers

- Increased back pressure and fuel penalty
- Lack of effective regeneration strategies to reduce input energy and deal with low exhaust temperature
- Durability of the system, including filter materials

Partners

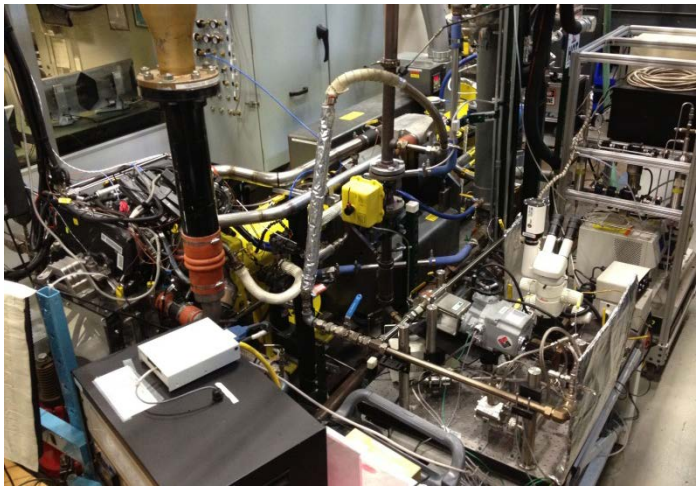
- Corning and Caterpillar
- University of Illinois – Chicago
- University of Wisconsin – Madison
- Tokyo Institute of Technology
- ILJIN Electric Co., Korea

Relevance and Objectives

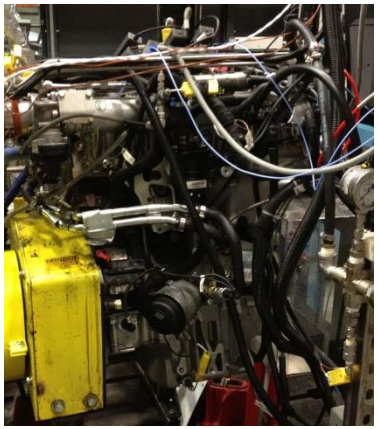
- Existing DPF systems still need to improve filtration/regeneration efficiencies and back pressure.
- DPF systems need efficient regeneration strategies, which can control thermal run-away.
- Low back-pressure filters are needed to be developed.

- Define soot oxidation characteristics with various gaseous emission compositions.
  - Provide soot oxidation kinetic parameters.
- Evaluate the effects of exhaust emissions on regeneration.
- Analyze the *μ*-pore structures of different filter materials to design the low back-pressure filters.
- Conduct numerical modeling to characterize the flow dynamics in DPF.

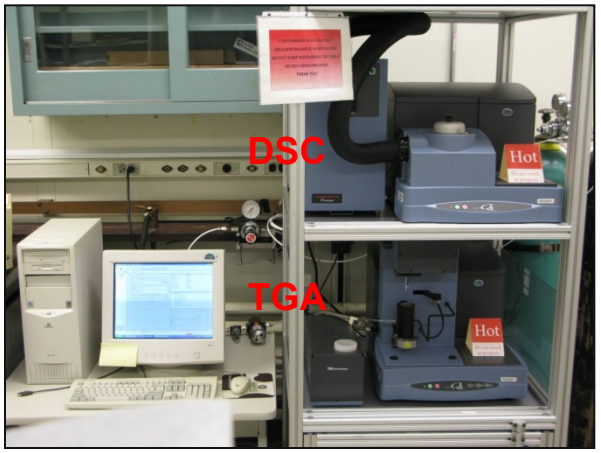
Approach



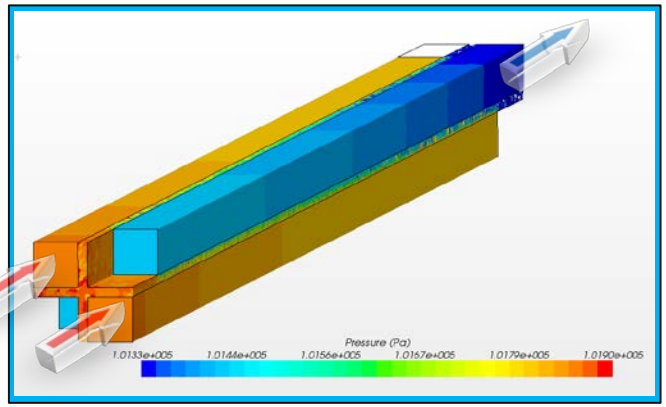
DPF experiments for filtration, regeneration, μ-imaging



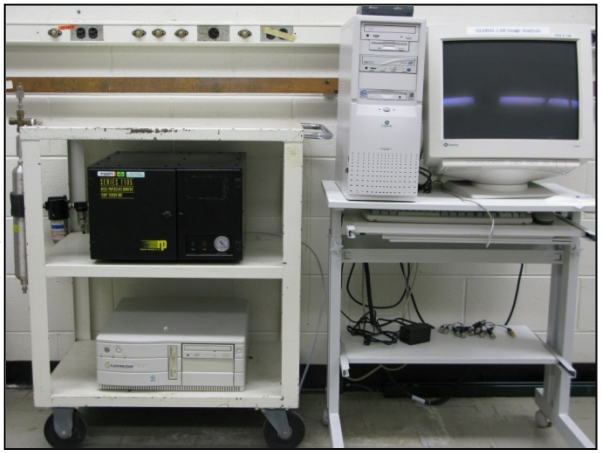
GM Diesel Engine



Soot oxidation experiments with TGA, DSC

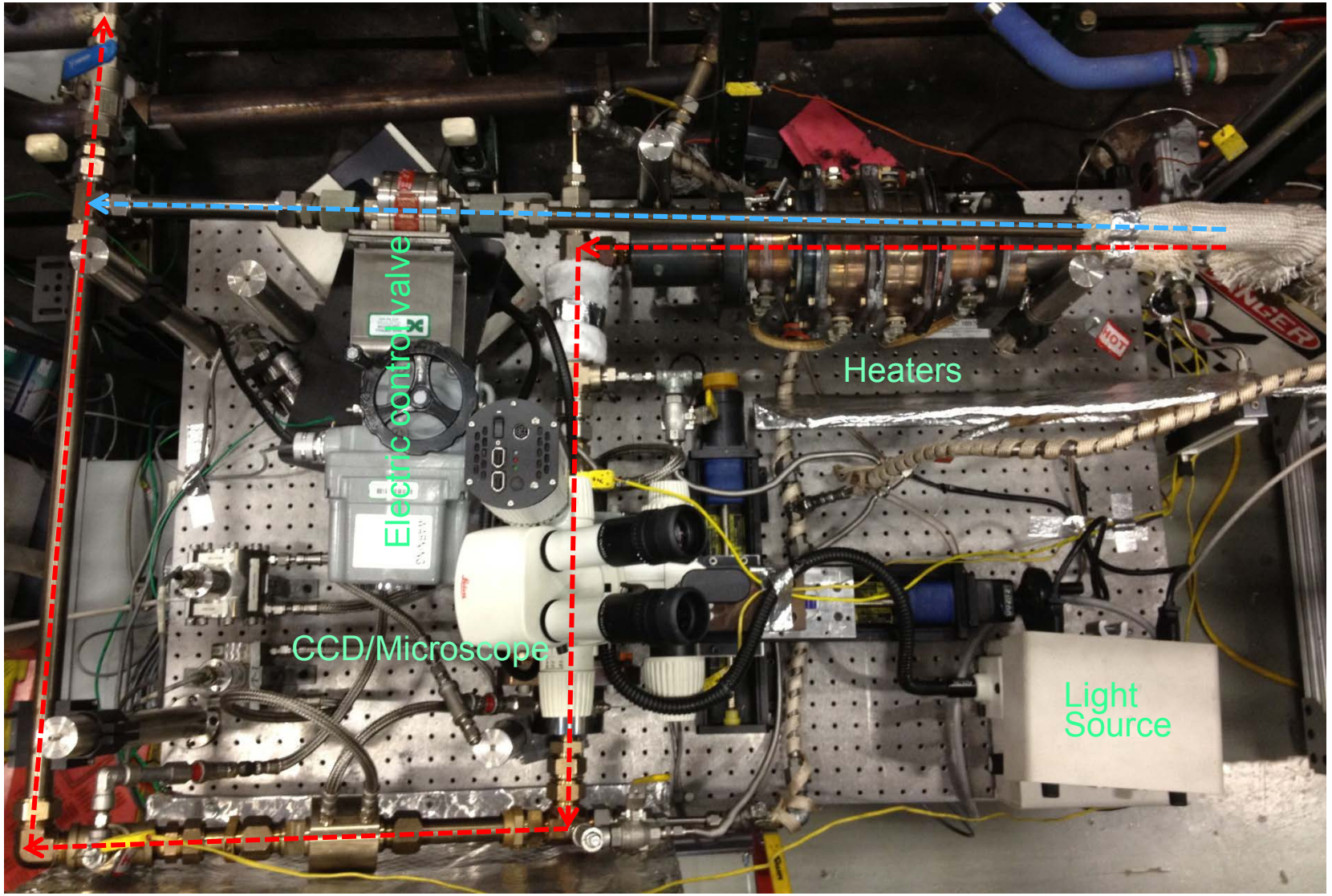


Numerical modeling



Soot mass, filtration efficiency with TEOM

DPF bench test system upgraded with automatic flow control



Most optimum experimental conditions and analytic methodology were defined for soot oxidation (FY 2010)

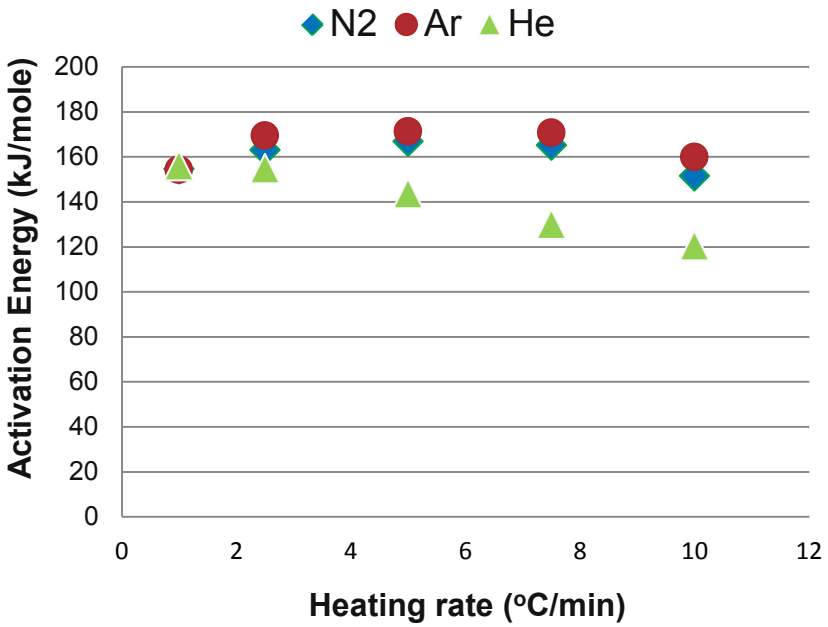
- Analytic methodology:

  Non-isothermal kinetic analysis using the differential method was proposed.

- Inert gas and heating rate:

  Inert gas-independent heating rate was found with 1 °C/min.

- Diffusion effects-independent experimental conditions were defined:
  - *Small sample (< 8 mg)*
  - *Shallow pan: Ø10 mm x H1.7 mm.*



$$\frac{d\alpha}{dT} = A\ exp\left(-\frac{E_a}{RT}\right) f(\alpha)$$

where $f(\alpha) = (1-\alpha)^n$

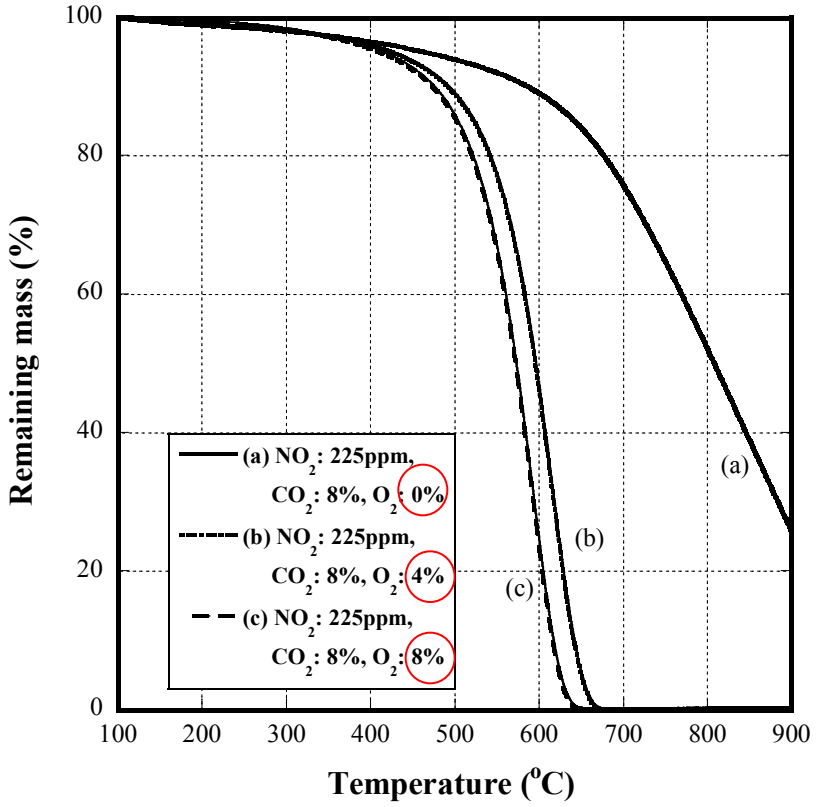
Soot oxidation experiments were performed with various reactant gas mixtures

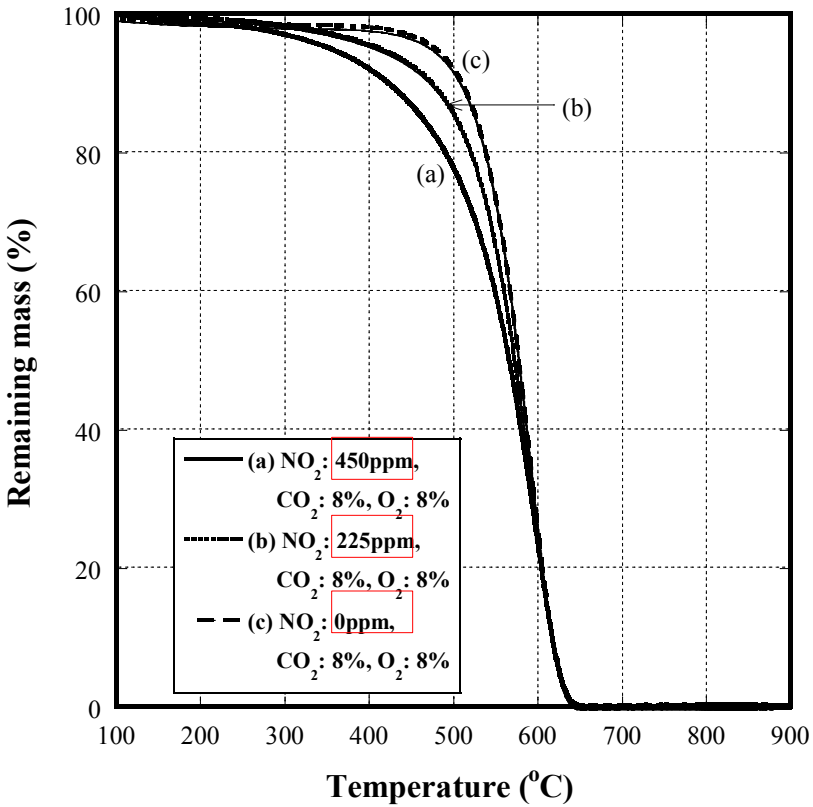
- Model soot: Carbon black (Printex-U)
- Instrument used
  - Thermogravimetric analyzer (TGA): To find oxidation behaviors and evaluate kinetic parameters (e.g., activation energy, reaction order etc.)
- Experimental conditions
  - Non-isothermal oxidation (50~900°C)
  - Initial soot sample mass:

    5.910 – 7.348 mg
  - Reacting gases: NO, NO_2, CO_2, O_2
  - Total gas flow rate: 100 ml/min
  - Heating rate: 1 °C/min

No	Concentrations of reactant gases			
	NO (ppm)	NO_2 (ppm)	CO_2 (%)	O_2 (%)
1	0	0	8	0
2	0	0	8	8
3	225	0	8	0
4	1000	0	0	0
5	0	225	8	0
6	0	225	8	4
7	0	225	8	8
8	0	225	0	8
9	0	180	3.5	13.5
10	0	450	8	8
11	0	1250	10	5.5

Both O_2 and NO_2 promote soot oxidation but in different temperature ranges

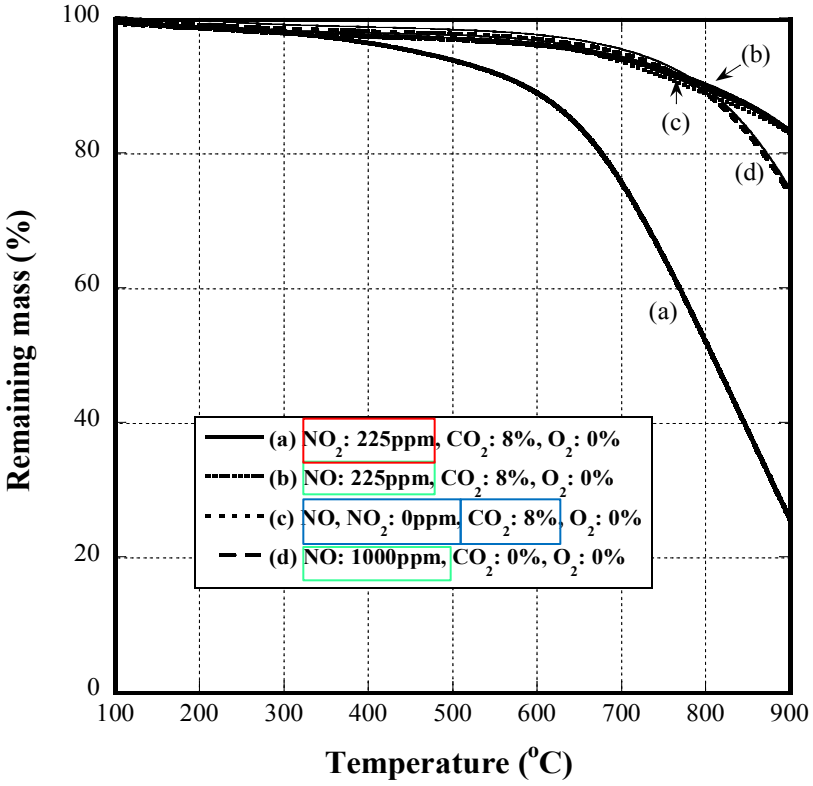


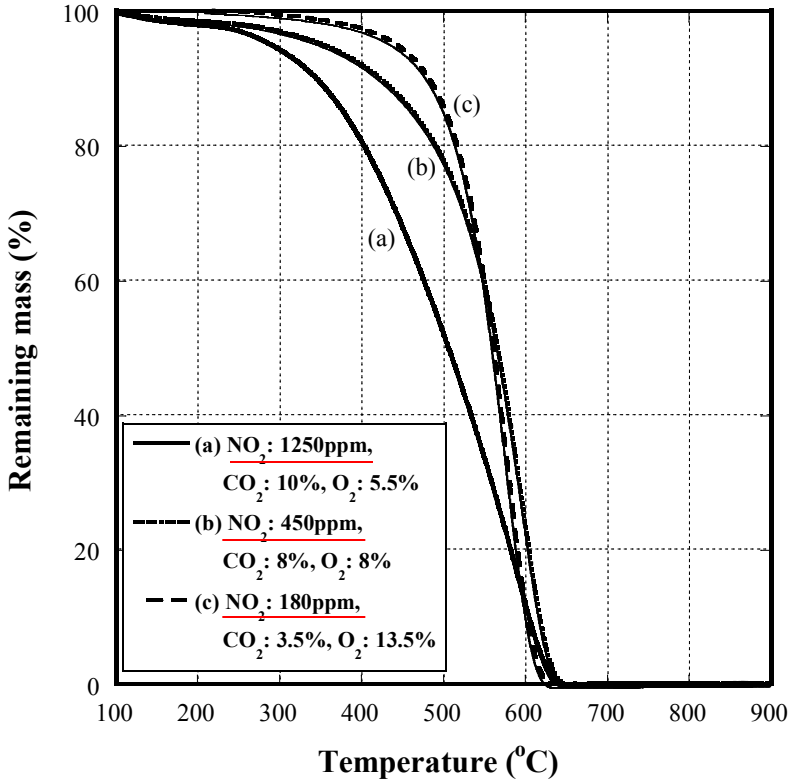


- Oxidation significantly increased from 0 to 4% O_2 ($\geq 400\ ^{\circ}C$)

- Oxidation range extended to the lower temp.
- Completion of oxidation at the same temp. was caused by the strong O_2 effect.

CO_2 and NO rarely affect soot oxidation. The high engine load condition promotes oxidation mainly by NO_2

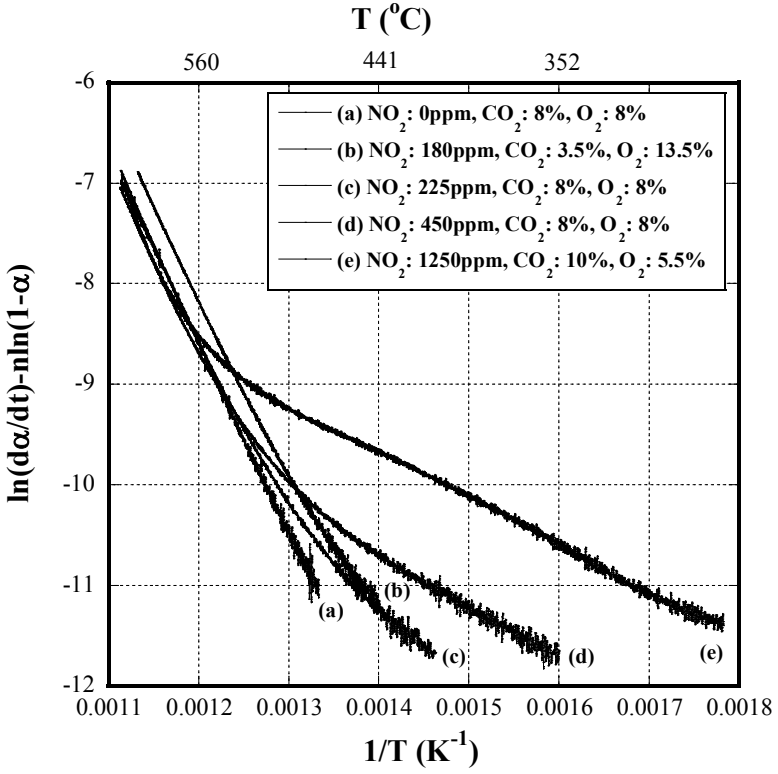


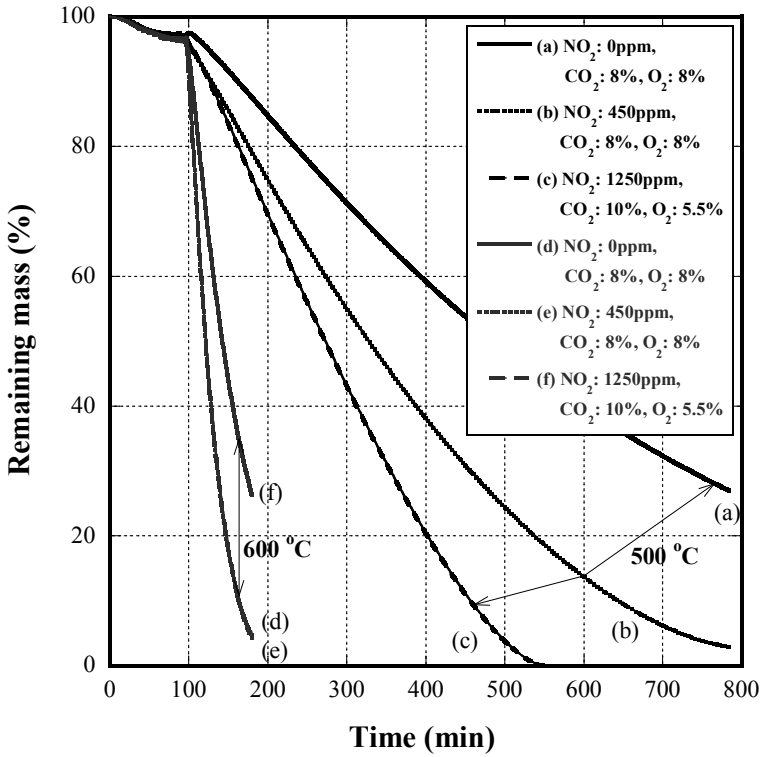


- Oxidation by CO_2 is quite minor.
- NO gas little affected soot oxidation.
- A little addition of NO_2 promoted oxidation significantly and shifted the starting point.

- (a) high (b) medium (c) low engine load conditions
- High NO_2 concentration at the high load condition (a) promoted oxidation by expanding the oxidation zone to the lower temperature (*potential thermal runaway control*)

Arrhenius plot shows two different oxidation zones with NO_2. Temperature dependence of NO_2 and O_2 was proven by isothermal experiments

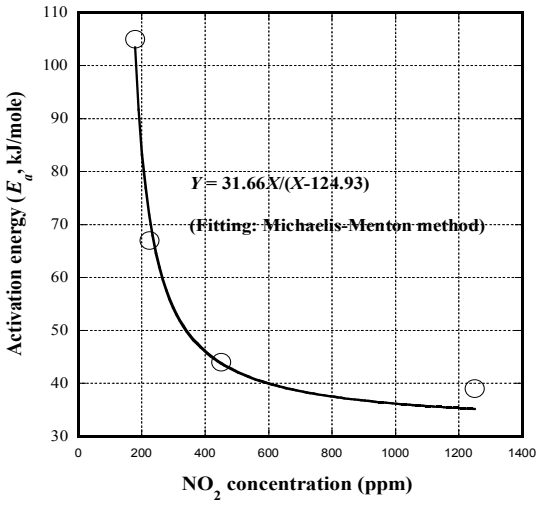




- Increased NO_2 concentration extended the low-temp oxidation.
- Two different sets of kinetic parameters are needed to be evaluated as NO_2 involved.
- At 500 °C, soot oxidation was significantly enhanced with an addition of NO_2.
- At 600 °C, the rate of soot oxidation appeared to be insensitive to NO_2 conc.

Activation energy was evaluated in two separate temperature regions

Compositions (NO_2: ppm, CO_2 & O_2: vol%)	Low temperature (LT) zone			High temperature (HT) zone		
	T (°C)	E_a (kJ/mole)	n	T (°C)	E_a (kJ/mole)	n
a) NO_2: 0, CO_2: 8, O_2: 8	T (°C): 477 – 625, E_a (kJ/mole): 153, n = 0.83					
b) NO_2: 180, CO_2: 3.5, O_2: 13.5	441 – 491	105	1	515 – 610	154	0.85
c) NO_2: 225, CO_2: 8, O_2: 8	410 – 471	67	1	526 – 625	157	0.80
d) NO_2: 450, CO_2: 8, O_2: 8	352 – 471	44	1	550 – 626	154	0.80
e) NO_2:1250, CO_2: 10, O_2: 5.5	288 – 500	39	1	575 – 616	159	0.85



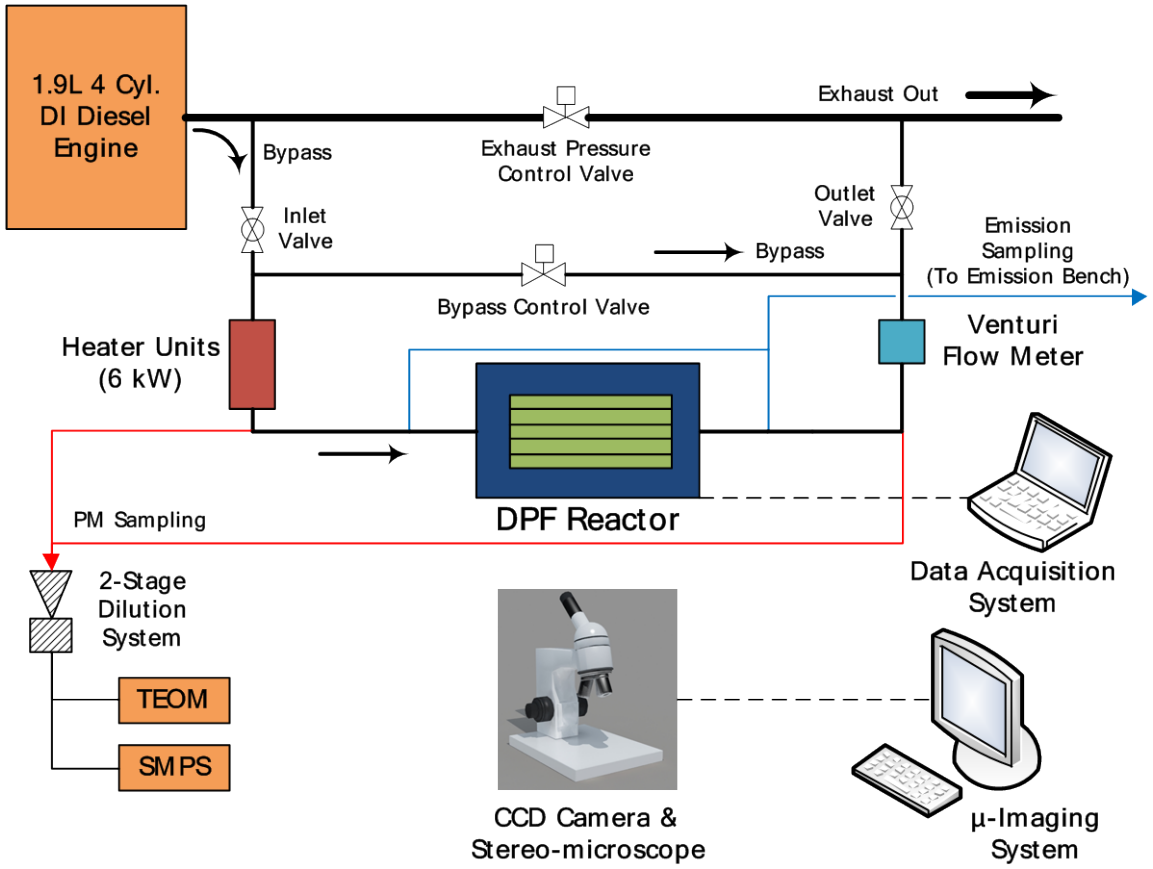
- E_a decreases as NO_2 concentration increases in the LT zone. → **Easier ignition of soot cake.**
- E_a is insensitive to gas compositions in the HT zone.
  → **Note the O_2 concentrations higher than 5.5%.**
- E_a's (averaged) from other literature [Jung et al.]
  - **0 NO_2, 5% O_2, 5% H_2O: 155 kJ/mole**
  - **900 ppm NO_2, 5% O_2: 62 kJ/mole**

- NO_2 effects on E_a (LT only) (typical diesel conditions)

$$E_a = 31.66\, C_{NO_2}\left(C_{NO_2} - 124.93\right)\frac{kJ}{mole}$$

Effects of NO_2 on DPF regeneration

- Experimental setup and conditions

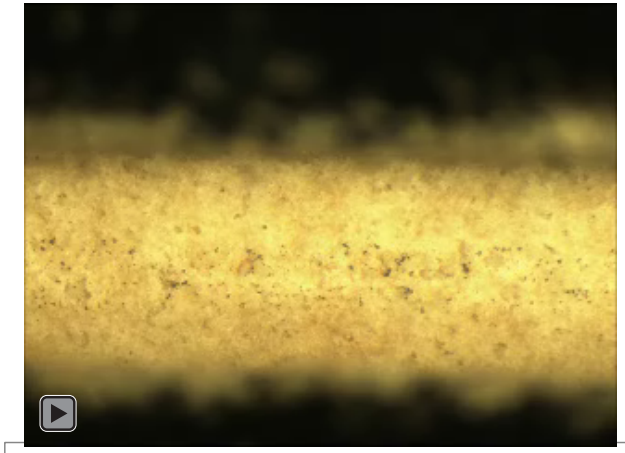


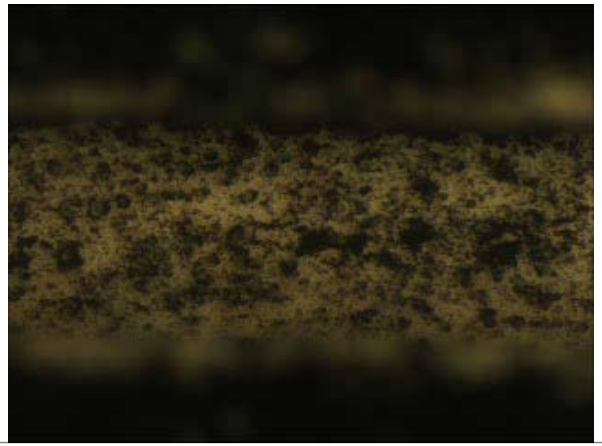
DPF Specification	
Size	2" x 6" half cut
Cell Density [cpsi]	200
Wall Thickness [in]	0.012
Porosity [%]	48
Mean Pore Size [micron]	13

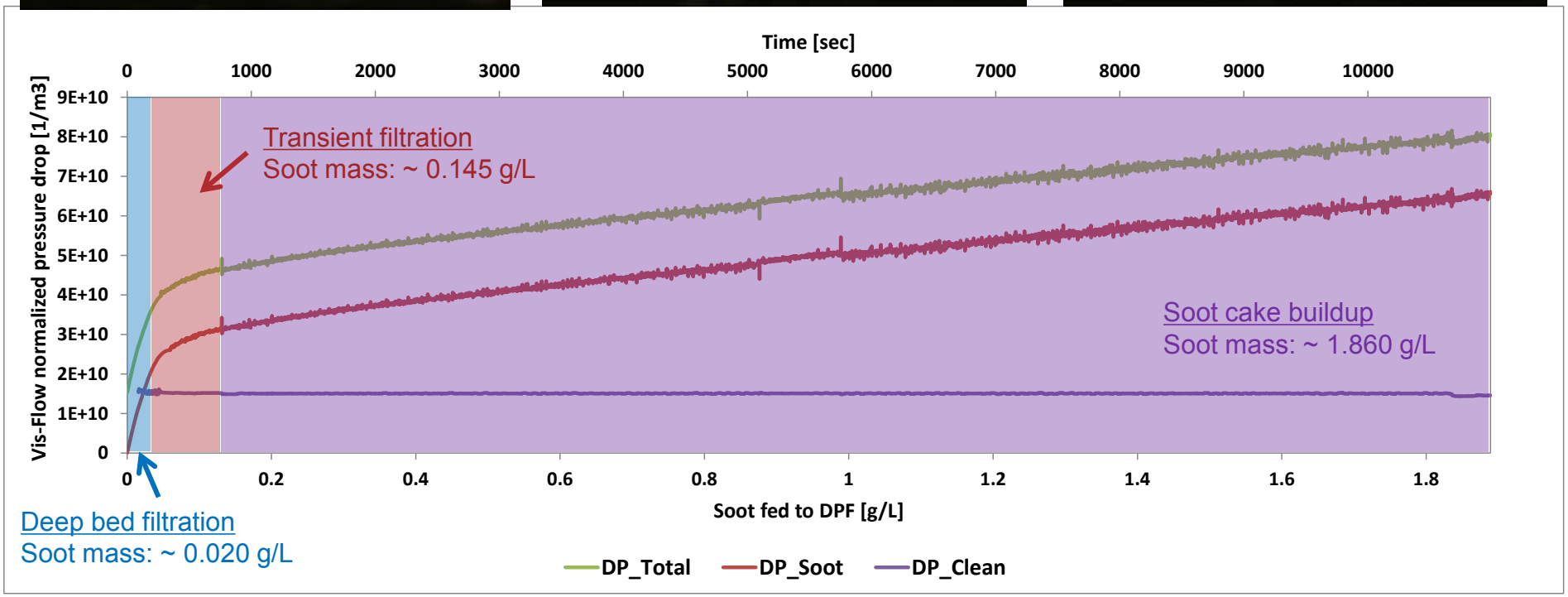
Engine Condition	
Speed [rpm]	2000
Load [bar BMEP]	4.7
A/F	23.75
Injection Timing [BTDC]	18 (PI) / 2 (MI)
Injection Pressure [bar]	930

Regeneration Condition		
O_2 Regen.	O_2 Conc. [vol%]	9.23 / 20.6
	Regen. Temp. [˚C]	500~540
	Wall Velocity [cm/s]	3.64
NO_2 Regen.	NO_2 Conc. [ppm]	1000
	O_2 Conc. [vol%]	20.6 (air)
	Regen. Temp. [˚C]	400 / 500
	Wall Velocity [cm/s]	3.79 / 4.18

Filtration for total soot loading 1.86 g/L

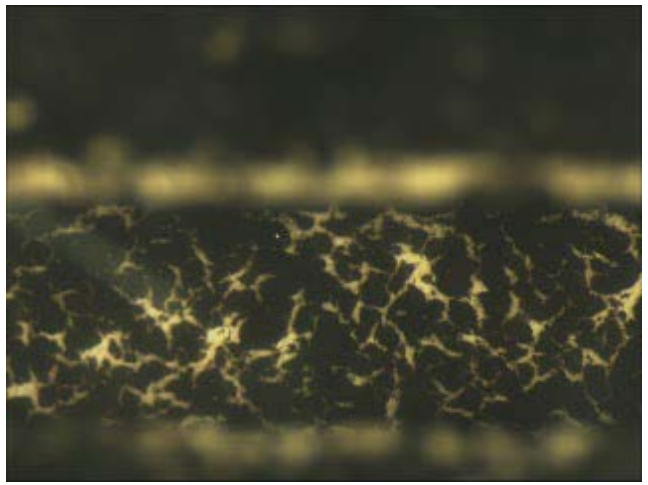


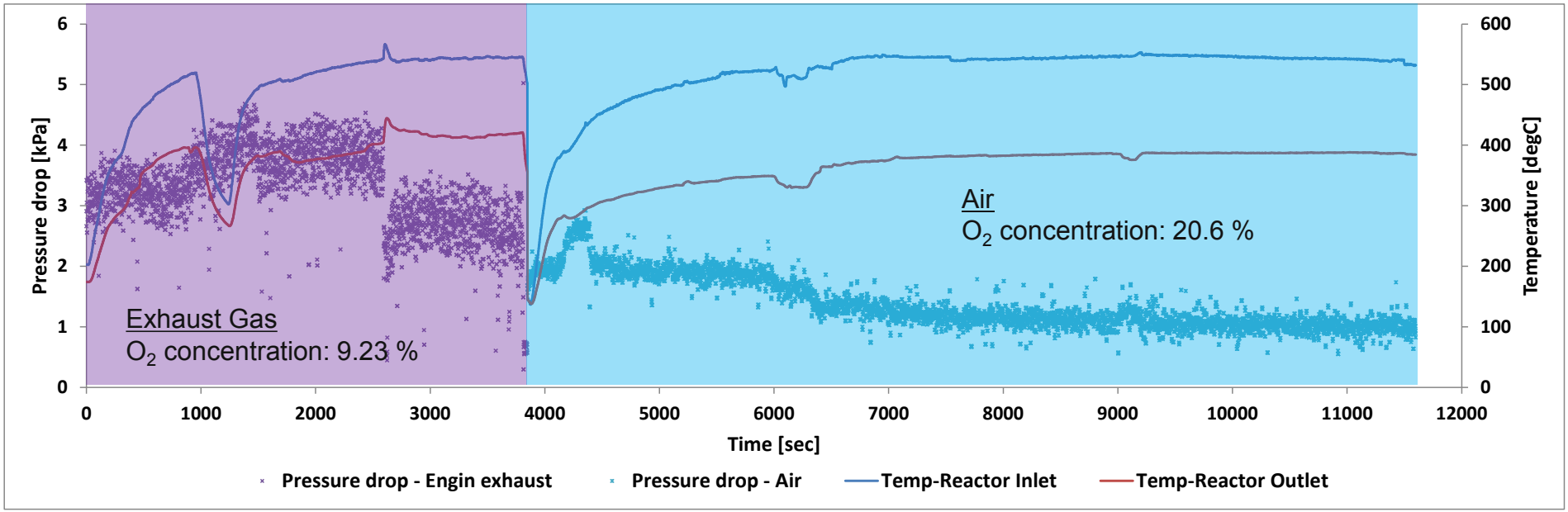




Regeneration with raw exhaust emissions and air without NO_2 assistant

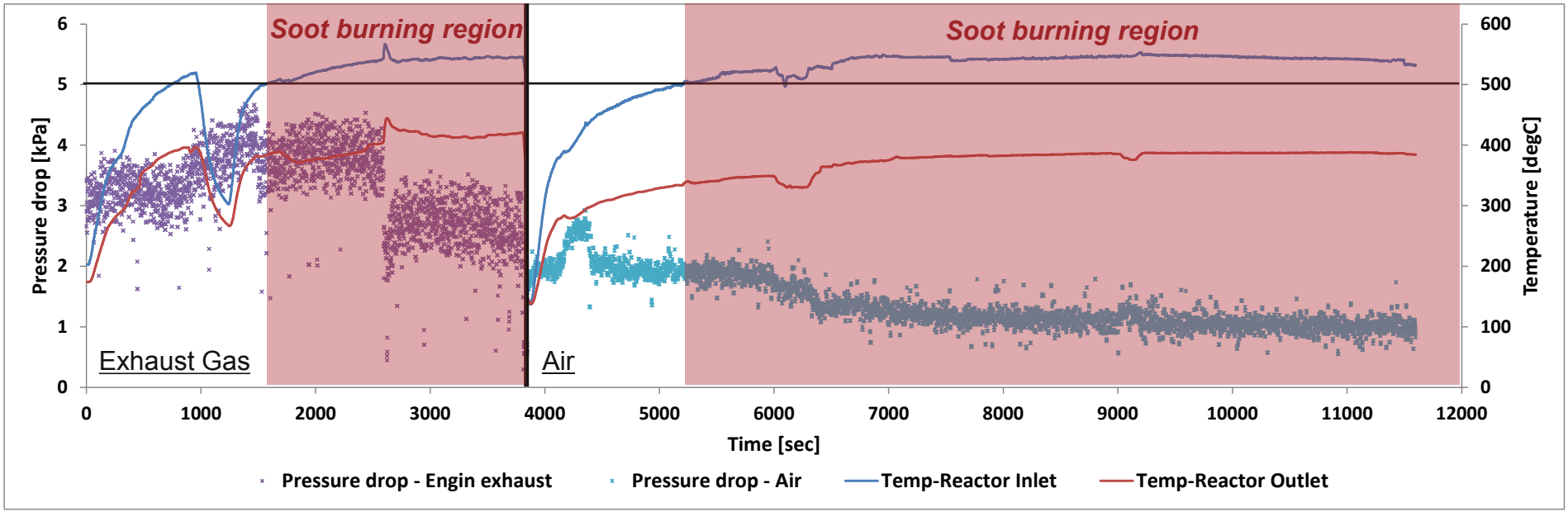
- Total soot loading: 1.86 g/L (287 mg)
- Regeneration temperature
  - Max. 540 ˚C
- Both engine exhaust gas (9.23 % O_2, 20 ppm NO_2) and air (20.6 % O_2) were used.



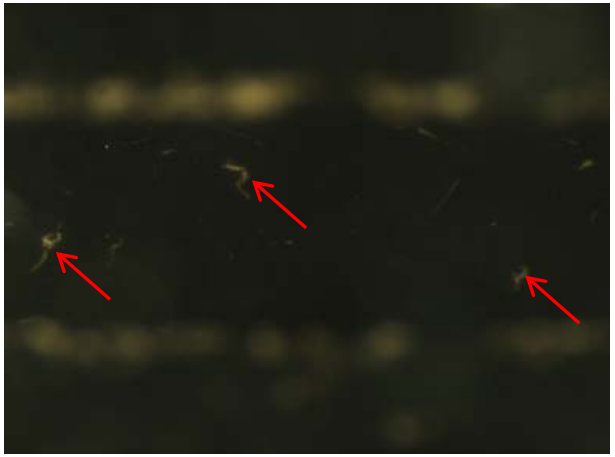


Major soot oxidation starts as inlet temp reaches 500˚C – No luminescent flame or flame propagation observed

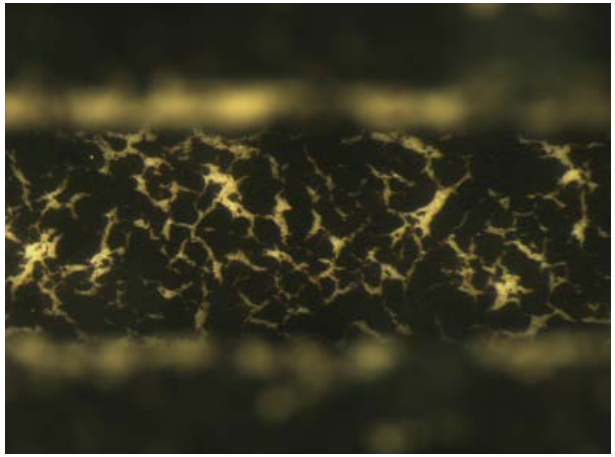
- As reactor inlet temperature is lower than 500˚C, the loaded soot hardly burns regardless of O_2 concentration.
- In 500 ~ 540˚C, it took more than 2 hours (8,200 sec) to burn out the 1.86 g/L (287 mg) soot deposit without assistance of NO_2.
  - The result agrees with TGA oxidation data tested at 500˚C (isothermal).
  - The oxidation of engine soot by O_2 gas alone proceeds at a very low rate.



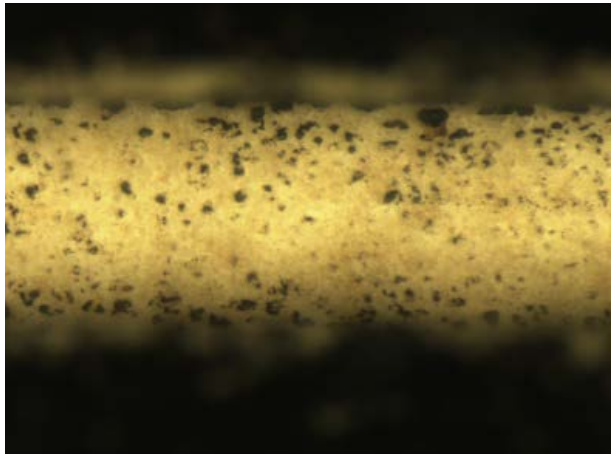
Soot in the deep bed is oxidized first, followed by a higher rate of soot oxidation on the channel, particularly near the opened pores



1,000 s after regen.



2,000 s

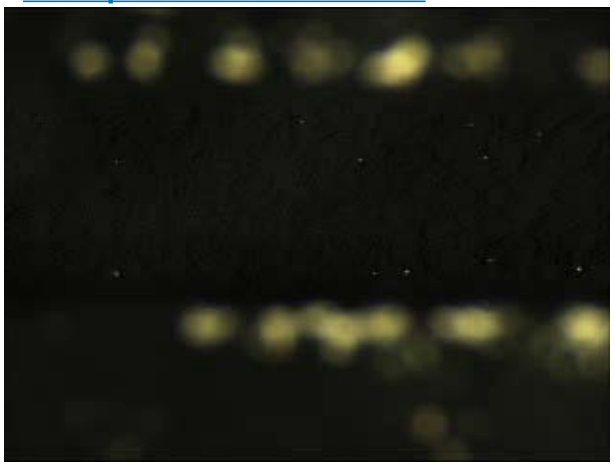


8,000 s

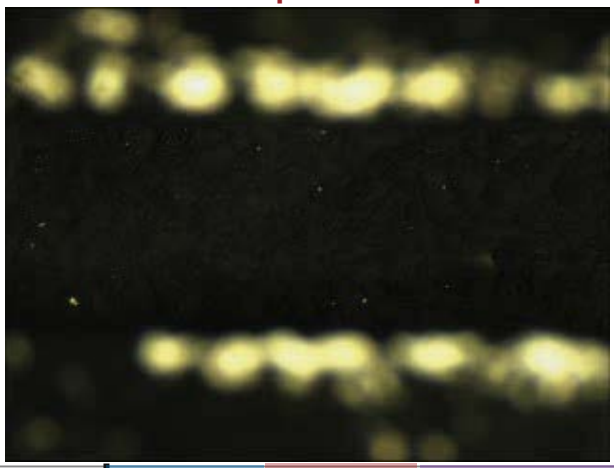
- Wall surface starts to be open in about 1,000 sec after regeneration (red arrows) → soot in opened pores seems to burn first.
- Soot in those surface areas early opened burns faster, due to a higher degree of diffusion of reactant gases.
- The soot cake continues to burn, but, at a relatively lower reaction rate.
- Some closed pores still exist with soot deposits until the last minute of regeneration (8,000 sec after regeneration).
- More closed pores appear near the edges of channel due to the lower oxidant flow rate.

NO_2 regeneration process was identified by analyzing μ-images and pressure drops in three reaction steps

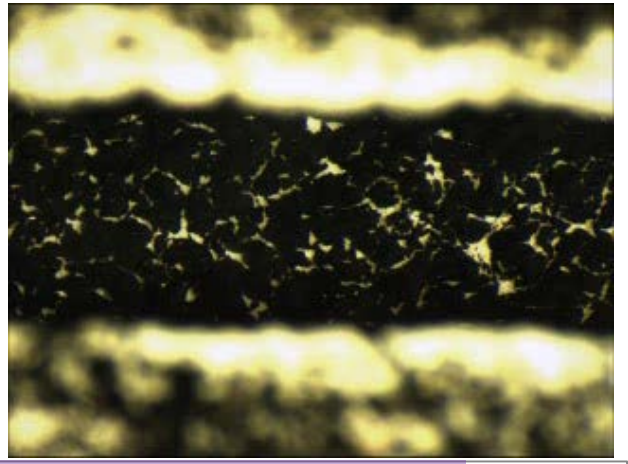
1st step:
Most pores are still closed



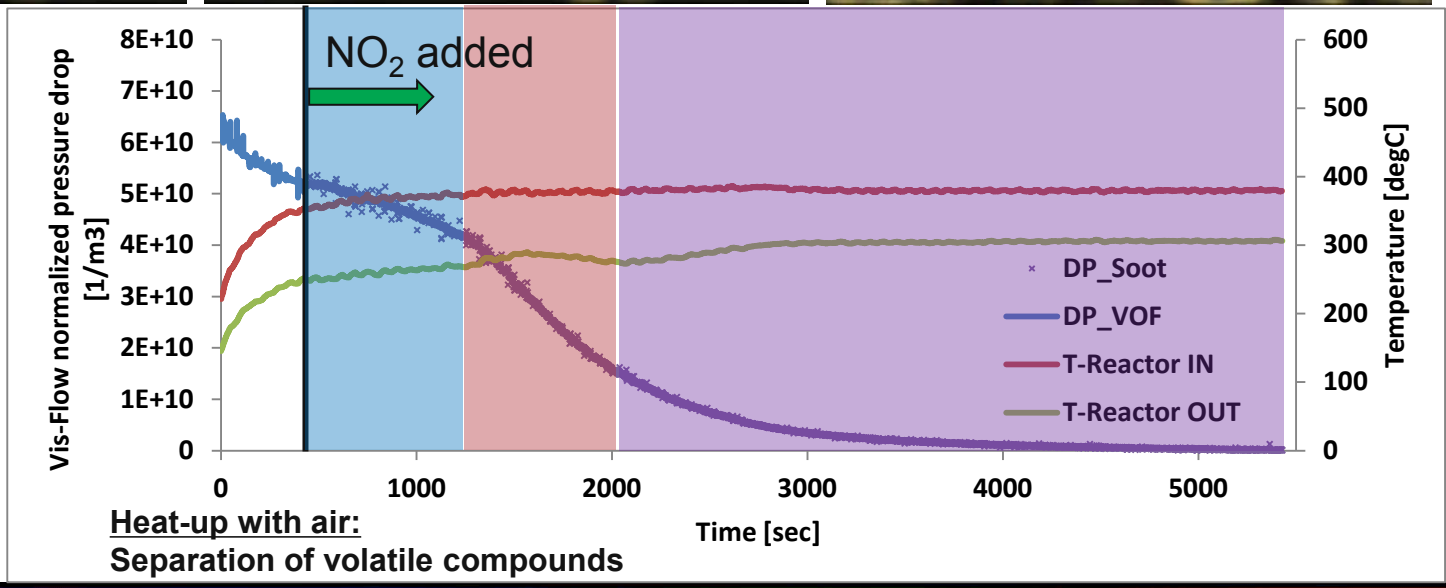
2nd step:
Soot oxidation in pores is completed



3rd step:
Slow oxidation on the soot bed



- Volatile compounds are evaporated during heat-up period. (about 25 % of total soot)
- During 1st step, soot in pores seems to be oxidized before pores are opened.
- Rapid pressure drop occurs in 2nd step, due to opened pores.

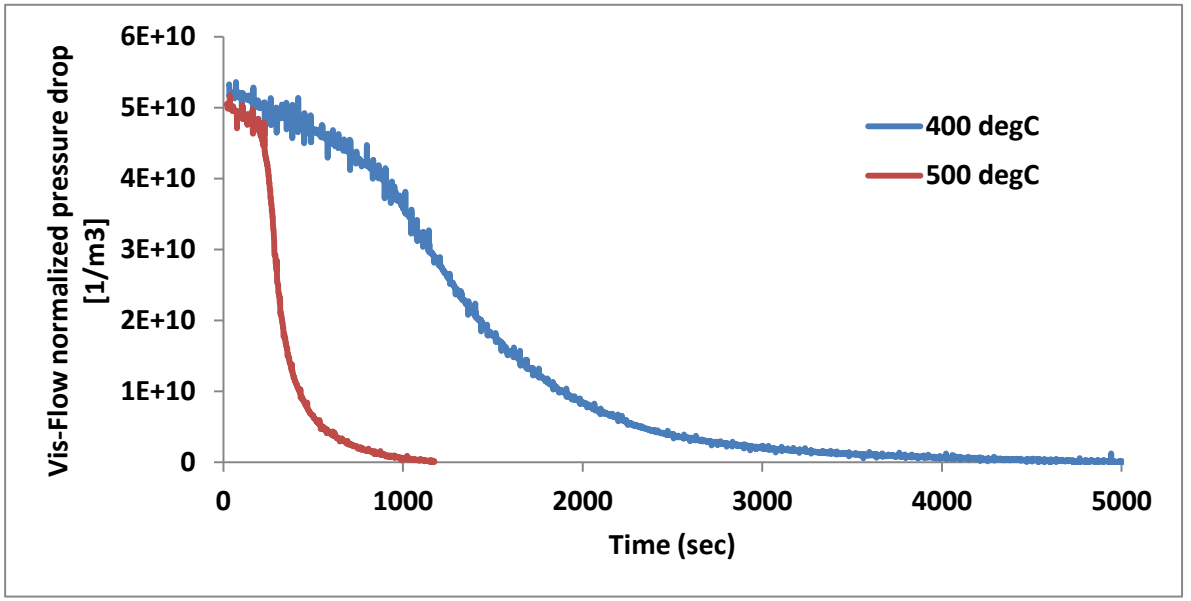


Heat-up with air:
Separation of volatile compounds

NO_2 promotes soot oxidation significantly

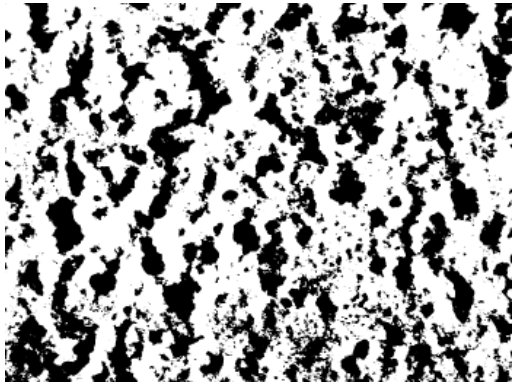
- To oxidize 1.86 g/L soot in DPF completely,
  - O_2 takes 8,200 sec at the inlet temperature 500 ~ 540 ˚C
  - 1,000 ppm NO_2 takes 1,200 sec at 500 ˚C and 5,000 s at 400 ˚C.
  - NO_2 promotes DPF soot oxidation about 7 times faster than does O_2 at the same inlet temperature.
- At the inlet temperature 400 ˚C, NO_2 still oxidizes soot successfully, but the reaction rate turns out to be about 4 times slower than at 500 ˚C.
- In the practical engine system, it is hard to achieve high NO_2 concentration and exhaust temperature → *catalyzed DPF and/or DOC are required*.

	400 °C	500 °C
1st step	780 s	200 s
2nd step	1,620 s	400 s
3rd step	5,000 s	1,200 s

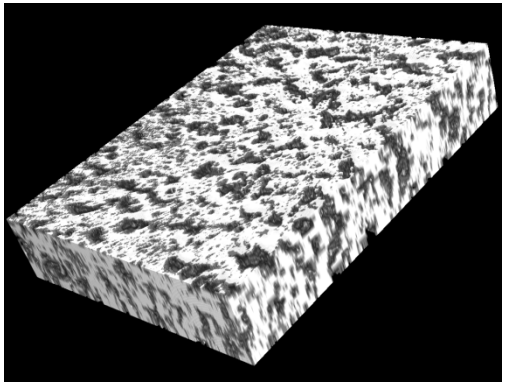


Filter μ-pore structures were imaged by conventional optics

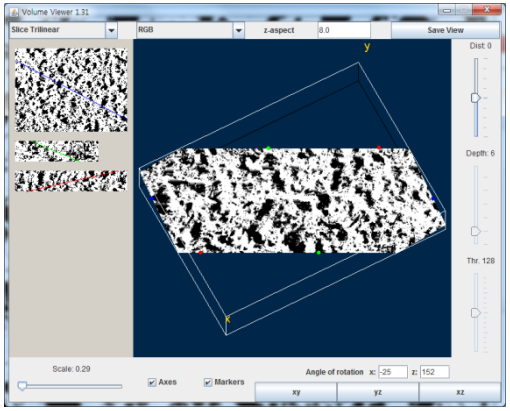
Depth	0 μm	80 μm	160 μm	240 μm
X 80	5~8 μm in depth			
X 400				



2-D pore image



3-D reconstruction



Sectional view using software

- Reconstructed 3-D images revealed the details of μ-pores at any section of filter web.

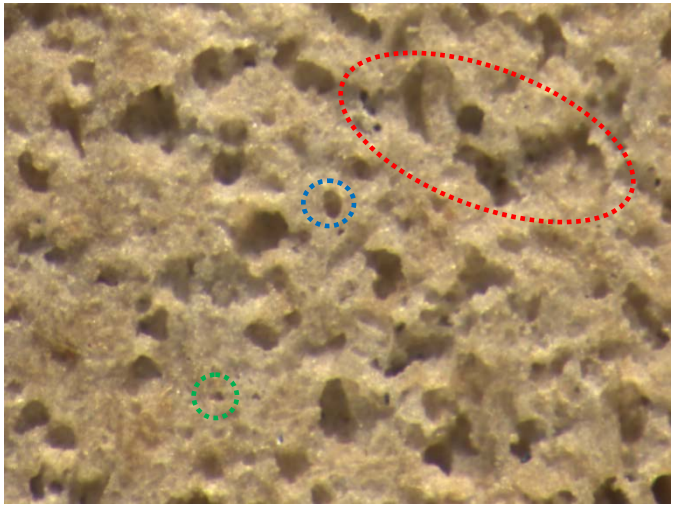
Pore structures were analyzed for various parameters

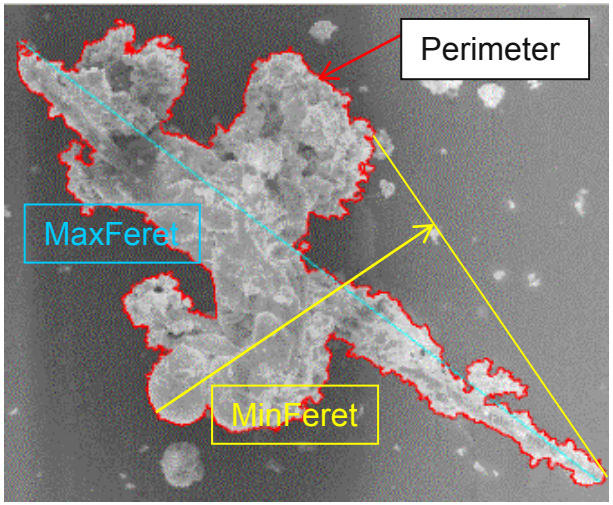
- Area of each pore and total area of pores
- Perineter: A total length of outer boundary of a selected pore.
- Circularity (C): $4\pi \times \frac{Area}{[Perimeter]^2}$
  - A magnitude of 1.0 indicates a perfect circle.
  - Decreasing circularity indicates a higher degree of elongation in shape.
- Feret's diameter: The distance between two parallel tangents of the two points on the pore boundary at an arbitrary angle.
- Feret ratio (FR): MaxFeret / MinFeret

Connected pore

Large single pore

Small pore

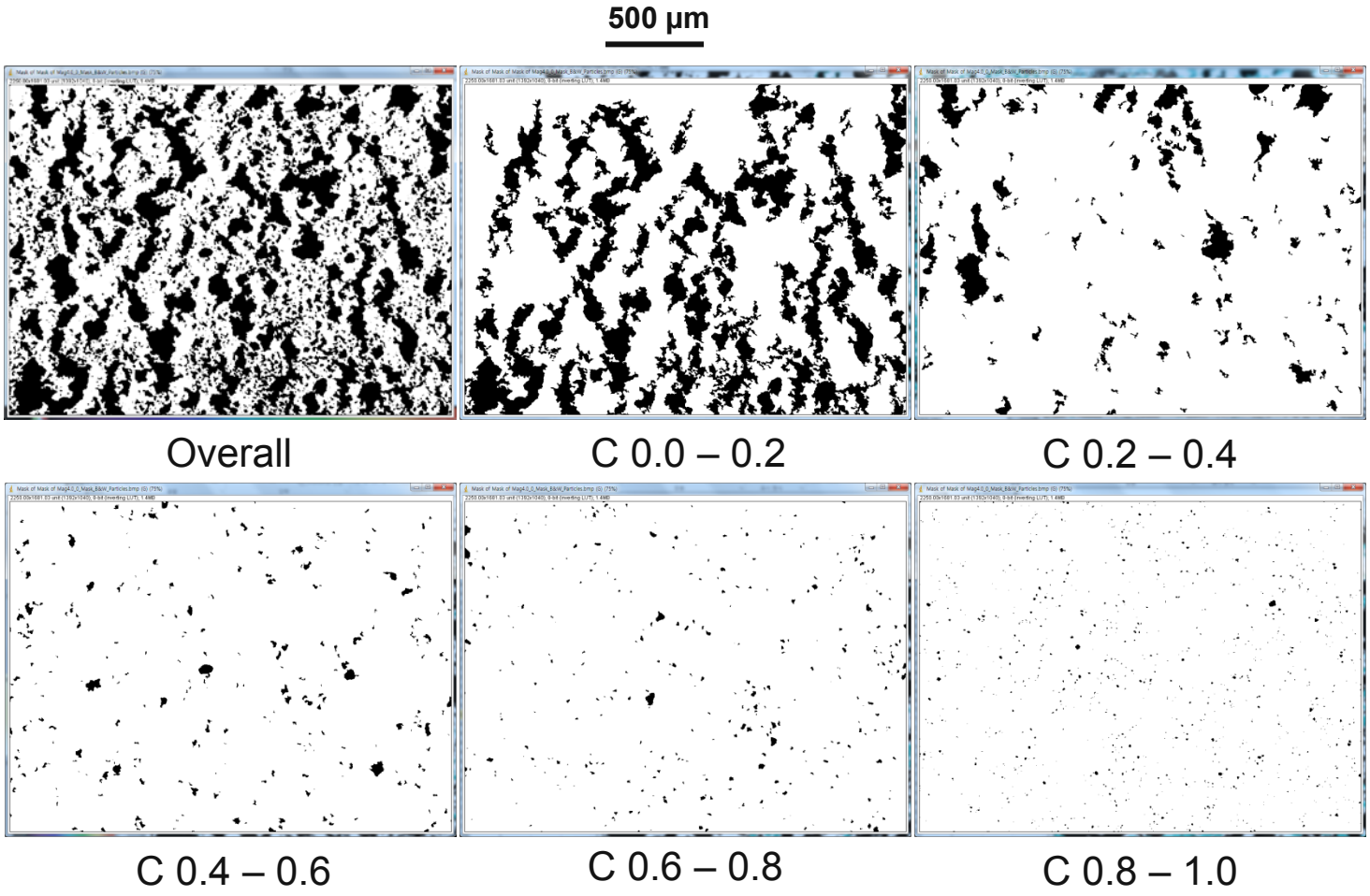




Pore geometry was analyzed for circularity and area

- As circularity (C) decreases, both pore size (area) and Feret ratio (FR) increase (elongated in shape)

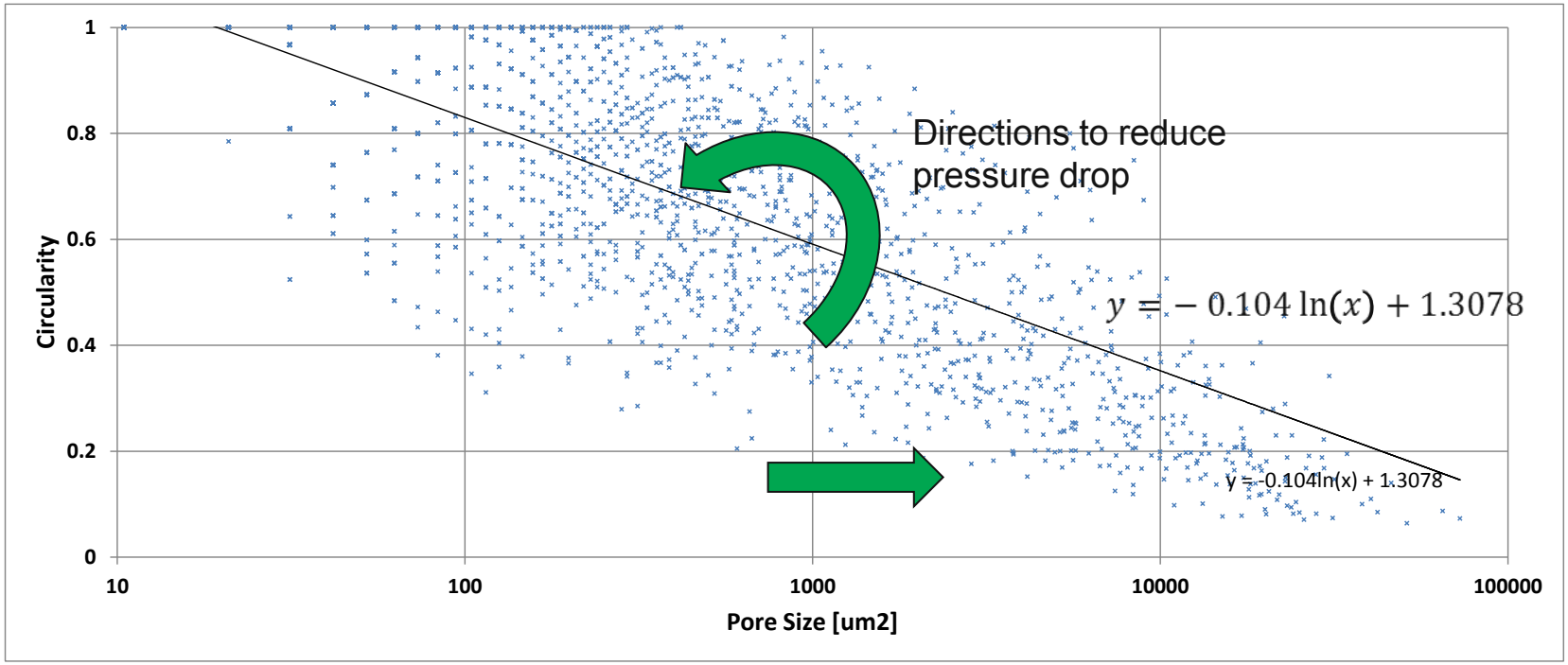
C	Size [um2]	FR
Overall	738.4	1.855
0.0-0.2	20554.4	2.155
0.2-0.4	2009.4	1.902
0.4-0.6	276.0	1.778
0.6-0.8	127.5	1.681
0.8-1.0	25.1	1.574



Parametric analysis proposes directions for filter pore design

- Circularity is inversely proportional to $log(pore\ area)$.
- As the *gradient* of linear regression line *increases*, the shape of connected or large pores becomes *more circular* and so the effective flow *area increases*.

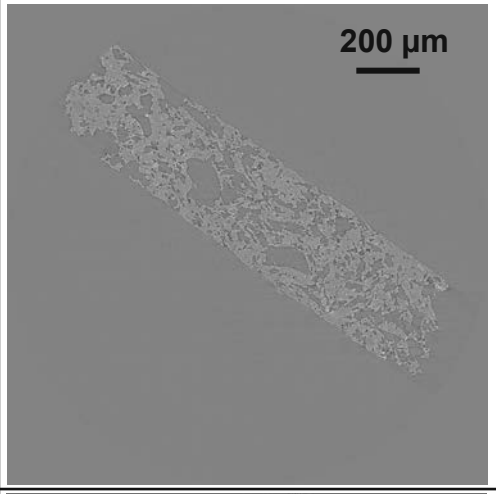
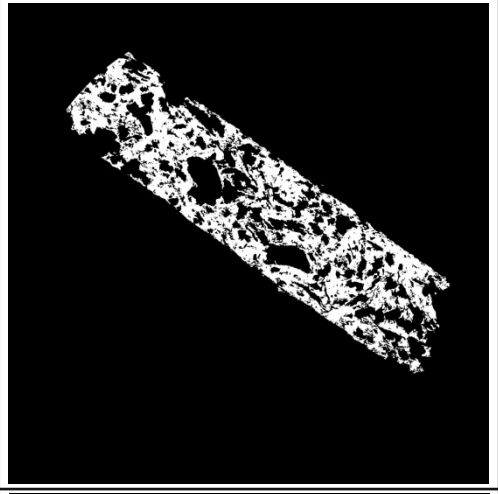
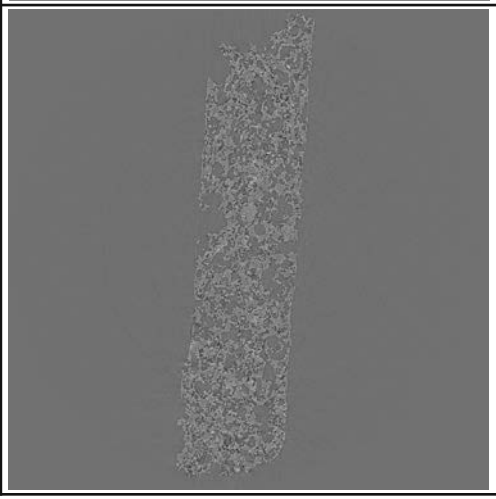
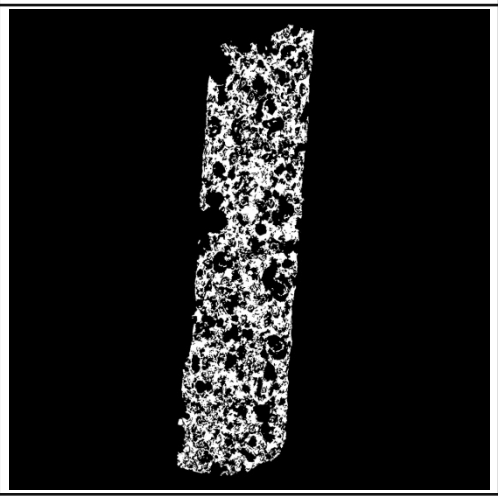
  → *Pressure drop will decrease*.



Circularity vs. Pore Size Correlation (analyzed for 2973 pores at X80)

X-ray microscopy visualizes further details in pore structures (using Advanced Photon Source)

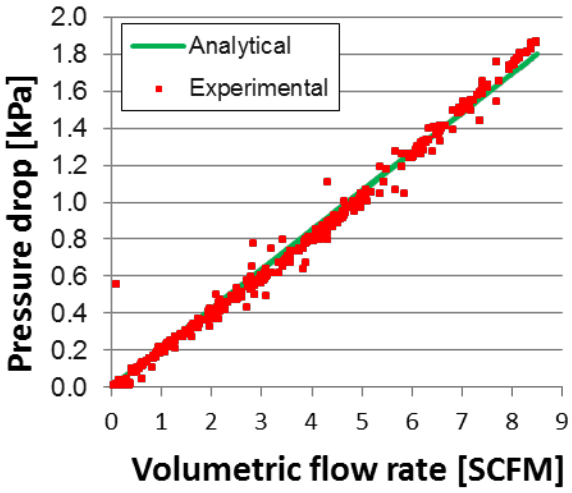
- Micro-tomography using X-ray attenuation images
  - Take images every 0.12˚ → 1500 images
  - Can analyze sectional images every 0.74 μm using tomography
  - Max resolution: 1 μm
- Image process
  - Raw image → Binary
  - Porosity has been calculated from the images
- Further capabilities
  - Pore size distributions
  - 3-D reconstruction
  - Pore path tracking

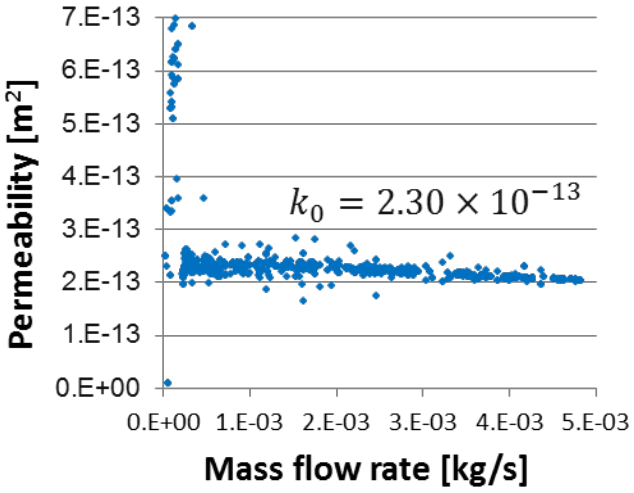
	Raw	Binary	Porosity
Filter (A)			44.89 %
Filter (B)			52.30 %

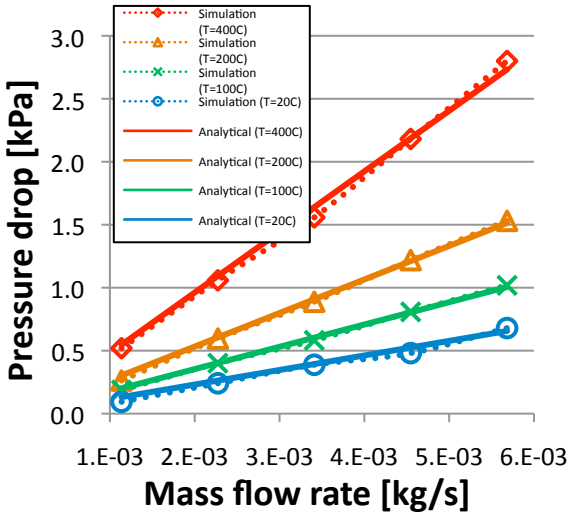
CFD Modeling of gas dynamics in DPF

- Assumptions
  - Steady state, Ideal gas (air), Laminar, Compressible flow
- Constants
  - Specific heat of air (1003.62 J/kg-K); Molecular weight of air (28.96 kg/kmol); Wall thermal conductivity (0.80 W/m-K); Wall porosity (0.45); Orthotropic wall flow; Density; Dynamic viscosity; Porous viscous resistance.
- Dimensions of sample filter
  - Cell density (200 cpsi); Cell size (H x W=0.0587” x 0.0587”); Channel length (6.0 in); Plug length (0.4 in); Web (wall) thickness (12 mils).
- Defining the volume mesh
  - Polyhedral : Avg. 14 faces
  - Mesh base size: 0.50 mm
  - Total # of cells : 395,982
- Initial/Boundary/Iteration conditions
  - Initial P_{inlet}, P_{outlet}: 101,325 Pa (1 atm)
  - Initial T_{filter}, T_{fluid} : 20 ^{o}C
  - Stop running when the variation of $\Delta P \leq 1.0$ Pa in 100 consecutive iterations

Numerical modeling has been validated in comparison with experimental and analytical data







$$\Delta P = \Delta P_{wall} + \Delta P_{friction} + \Delta P_{cont./expansion} + \Delta P_{soot}$$

Negligible (cont./expansion) — Clean filter (soot)

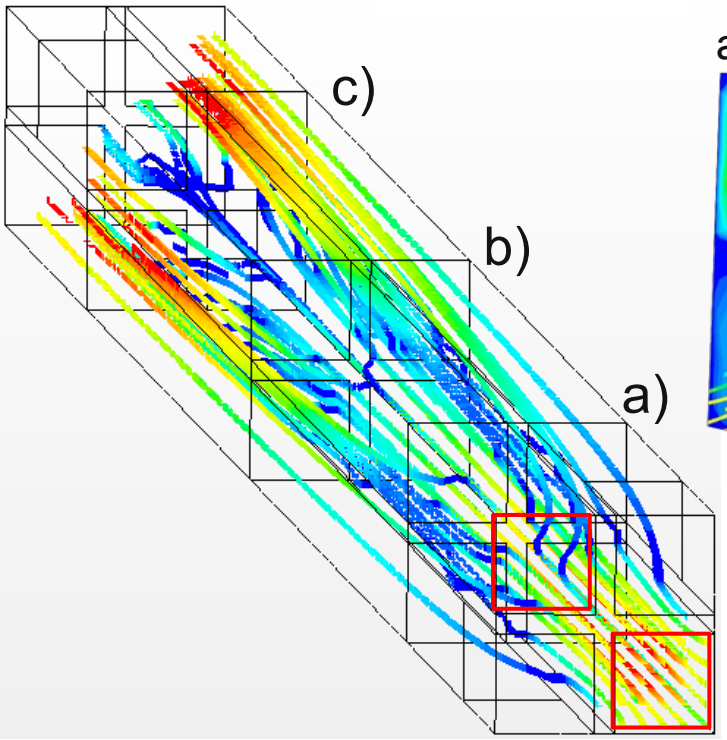
$$= \left(\frac{\mu}{k_o} u_w w_s + \beta \rho u_w^2 w_s\right) + \left(\frac{2\mu F}{3a^2} UL\xi\right)$$

Negligible ($\beta \rho u_w^2 w_s$)

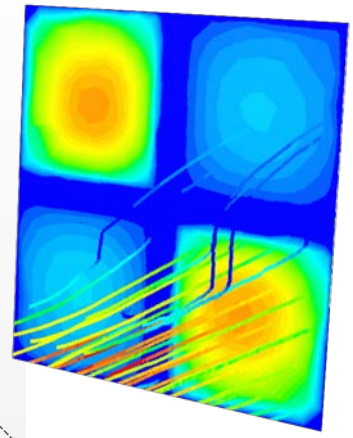
- Pressure drops calculated by the theory agree with experimental data quite well.
- Permeability of sample filter has been evaluated experimentally ($k_0 = 2.30 \times 10^{-13}\ m^2$).
- Pressure drops calculated by modeling agree with analytical data as well.

3-D gas velocity profiles have been calculated successfully

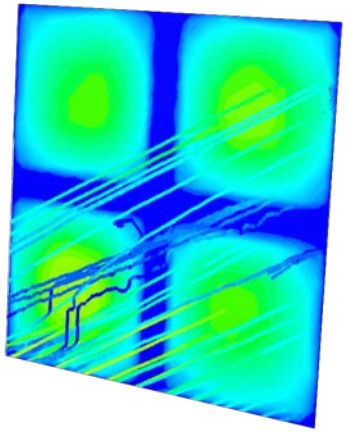
$$\dot{Q} = 8\ scfm$$



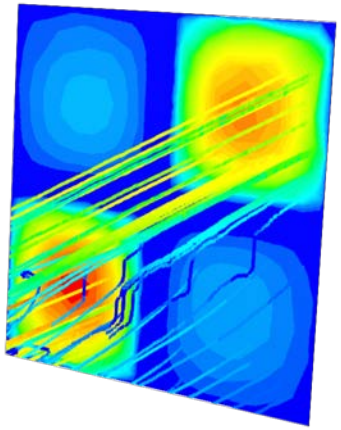
a) 1” from inlet

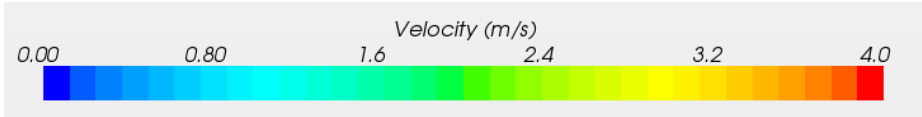


b) 3” from inlet



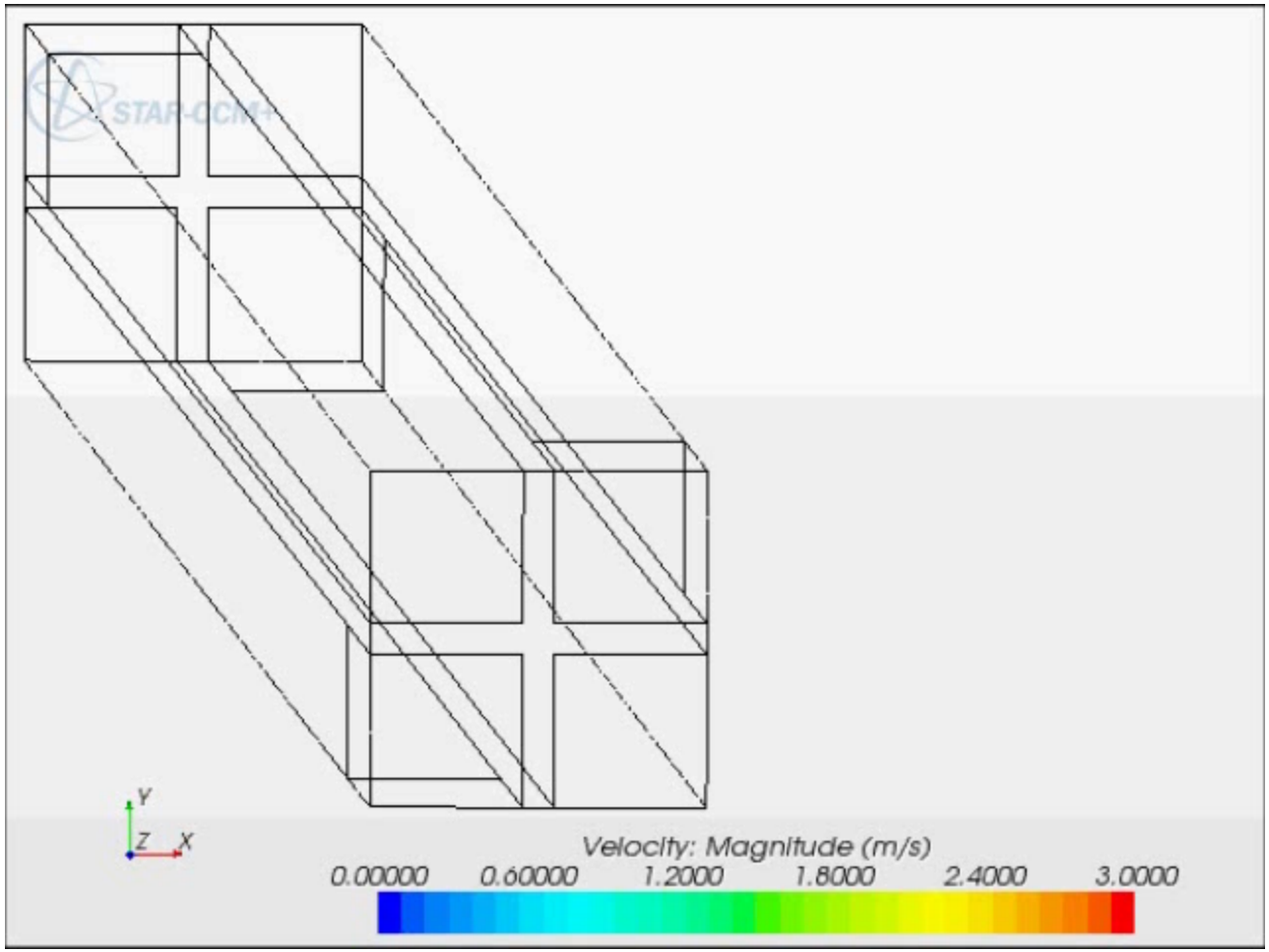
c) 5” from inlet





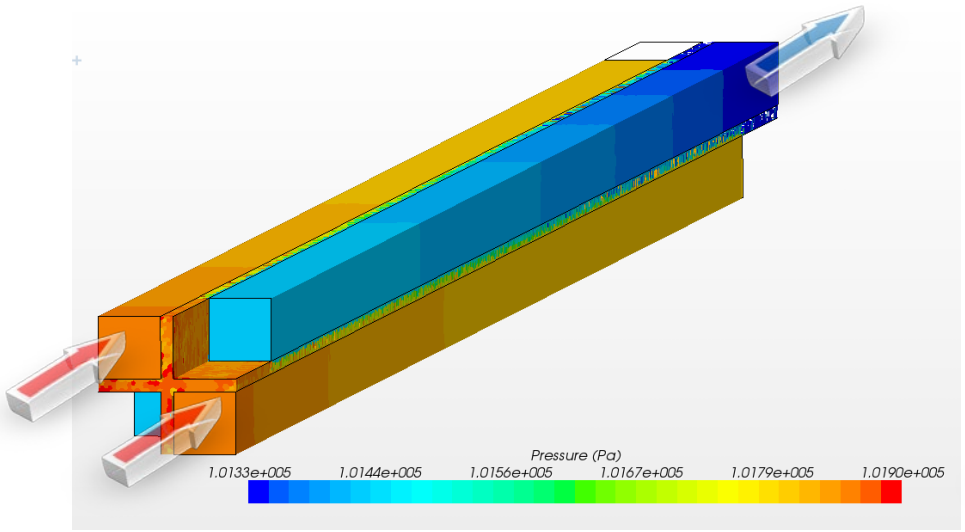
Incoming channels (2)

3-D motion picture shows actual gas flow dynamics with velocity distributions

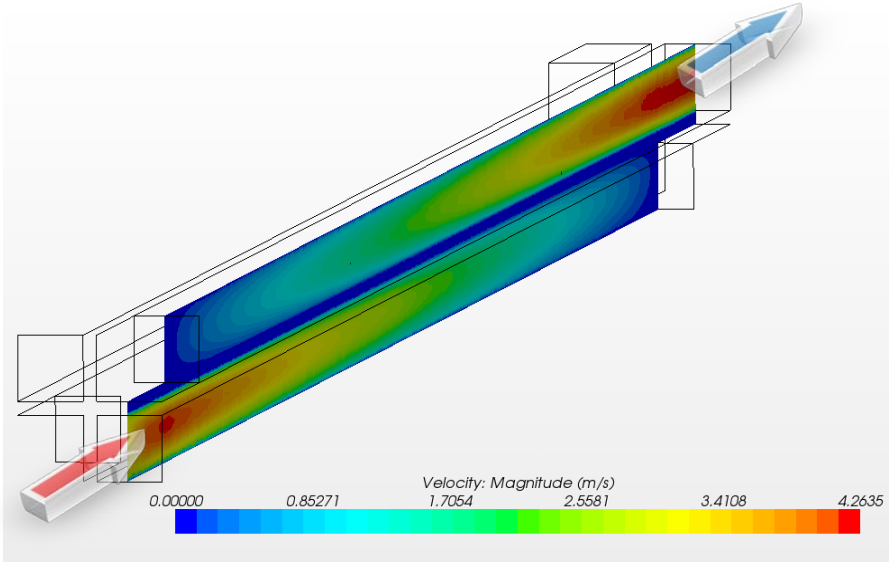


Pressure and velocity distributions have been calculated along the channels and on a cross sectional area respectively

$$\dot{Q} = 8\ scfm$$

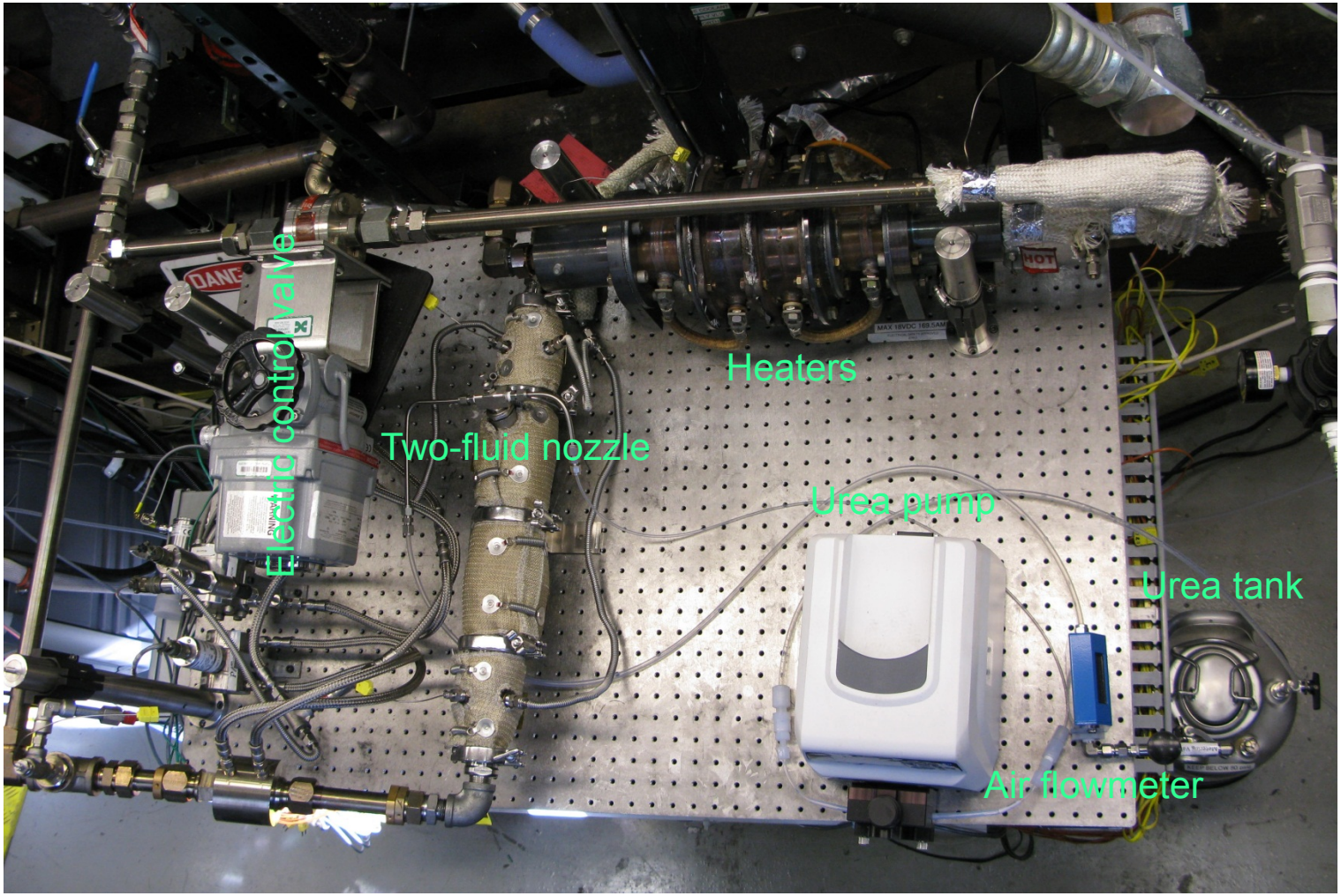


Pressure distributions



Velocity distribution

A bench-scaled urea-SCR test system has been built for future use



Future Work

- A new CRADA project will start in 2012 (a contract in progress).
  - Title: *Particulate Emissions Control by Advanced Filtration Systems for Light-duty Engines*
  - Sponsors: DOE office of VT, Corning, Inc., and Hyundai Motor Co.
  - Scope of work (4 major topics)
    - *Catalyst coating effects on (conventional) particulate filter performance.*
    - *Performance evaluation of direct soot oxidation catalysts (DSOC).*
    - *Evaluation of engine soot oxidation behaviors (using TGA, DSC)*
    - *PM emissions characterization in size, morphology, nano-structures*

    ✓ *Deliverables: filter microstructures, soot cake properties (e.g., packing density, profile), regeneration performance (starting point, pressure drops, temperature distributions, and thermal runaway management), regeneration imaging, ash properties, aging effects.*

    ✓ *Variables: filter materials (AT, AC and others), catalyst chemical properties, amount of catalyst, engine operating conditions, gaseous emission compositions, inlet gas temperature*

- Numerical simulations will continue to be performed to study flow dynamics with soot loading and regeneration processes with chemical kinetics.

- The bench-scaled urea-SCR test system will be used to perform parametric studies that can find correlations with DPF filtration/regeneration processes.

Summary

- NO_2 and O_2 in exhaust emissions are major chemical components involved in soot oxidation. NO_2 promotes soot oxidation mainly in low temperature region and O_2 does in high temperature region, respectively.
- NO and CO_2 rarely contribute to soot oxidation.
- Activation energies for soot oxidation in NO_2–containing mixtures were first evaluated at two different temperature regions.
- The DPF regeneration experiments verified that the higher NO_2 concentration promoted soot oxidation. However, the catalyzed DPFs are necessary to be developed to improve DPF regeneration.
- The analysis for DPF μ-pores using the conventional optics offered a strong potential to design an advanced DPF membrane. The X-ray imaging technique provided an extremely high resolution of tomographic images.
- The numerical modeling showed a capability to define the 3-D gas dynamics in DPF and a strong potential to further characterize the fluid dynamics of soot-laden flow and the chemical reactions in DPF regeneration.
- The bench-scaled urea-SCR test system will provide many pieces of important information about its effects on DPF filtration and regeneration processes.

Accomplishment

- Publications
  - "Characterization of Oxidation Behaviors and Chemical-Kinetics Parameters of Diesel Particulates Relevant to DPF Regeneration," SAE 2010-01-2166, Oct. 2010.
  - "Measurements of Heat Release of Diesel PM for Advanced Thermal Management Strategies for DPF Regeneration," Journal of Combustion Science and Technology, 183:1, 2011.
  - During the entire project period (total 13+ papers)
    - *2 journal papers*
    - *8 conference proceedings published*
    - *1 journal and 2 conference papers to be published in 2012*
- Patents and Inventions
  - One (1) invention filed and one (1) invention ready to file
- A complete set of bench-scaled DPF test system has been fabricated, fully integrated with a heater system and measurement instruments, such as TEOM, SMPS, and emissions bench.
- A bench-scaled urea-SCR test system has been fabricated, which can be integrated with the DPF test system.
- Capability in numerical modeling has been developed for DPFs.

Contact

Kyeong Lee

Argonne National Laboratory

– (630)252-9403

– *klee@anl.gov*