Soot Deposits, Catalyst Coatings and Ash Layers: Creation, Function and Fate in Diesel Particulate Filters

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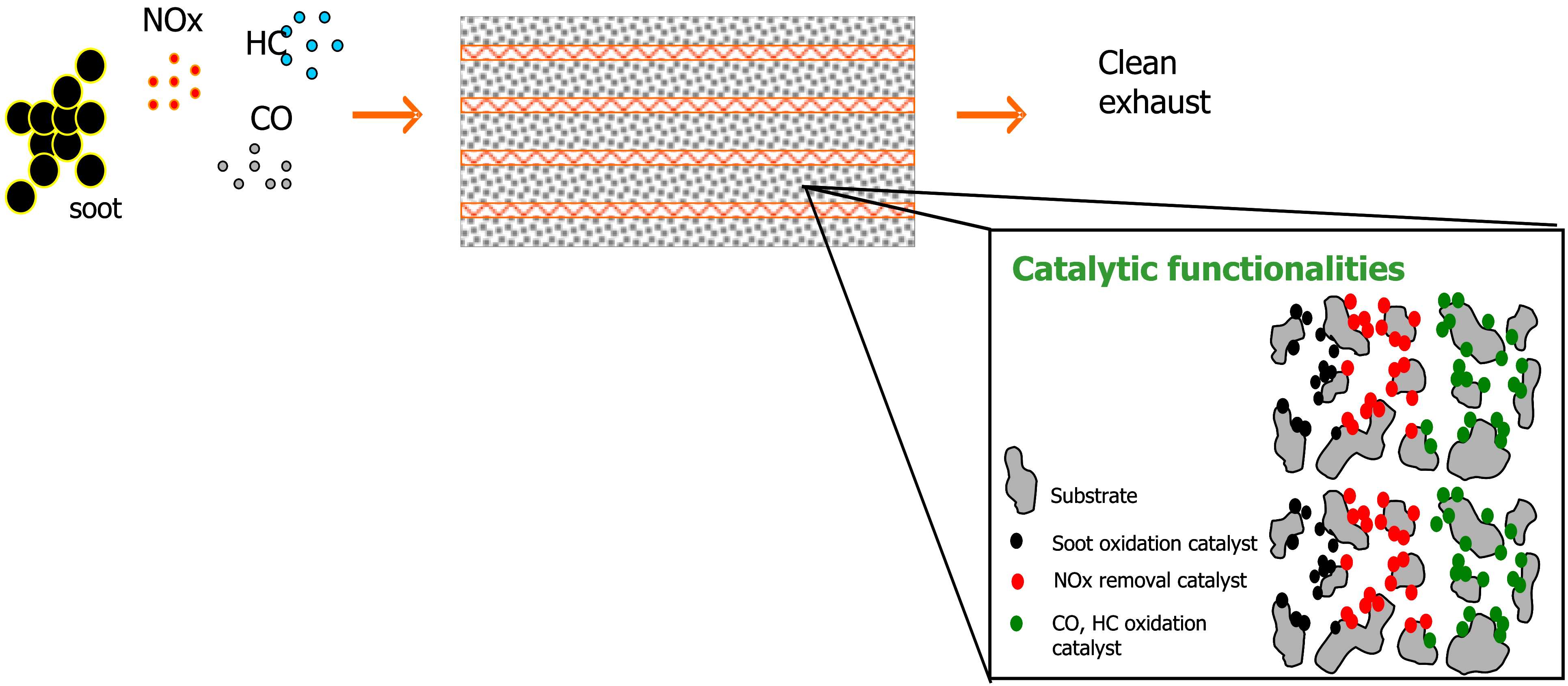
Trends in OEM DPF Functionalities

Integration/Compactness/Cost Reduction/Energy Efficiency



Back to Basics: The Actors

- **Soot deposits**

  Creation: microstructure (porosity) vs soot aggregate structure (fractal dimension) and deposition conditions (Peclet number, compaction)
  Function: flow resistance (permeability)
  Fate: reactivity (soot oxidation), "next day" effects (deposit restructuring)

- **Catalyst coatings**

  Creation: chemical composition/synthesis & coating technologies
  Function: direct (microstructure vs. soot-catalyst contact) and indirect (NO_2-assisted oxidation), gas species (CO/HC) oxidation, NOx control
  Fate: aging (e.g. thermal), ash induced impact

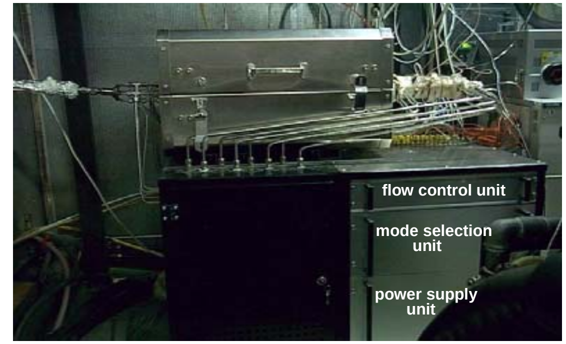
- **Ash Layers**

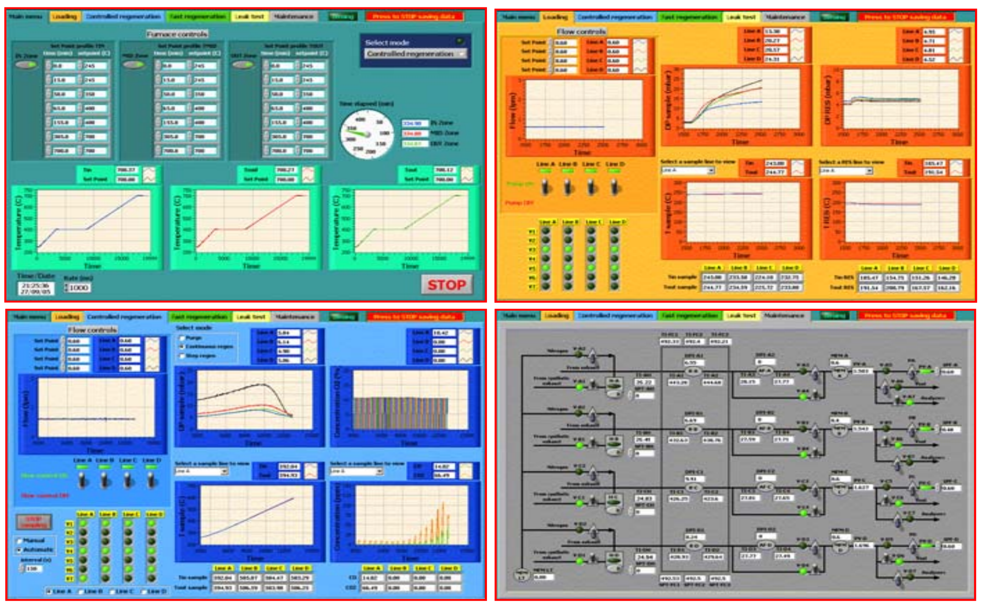
  Creation: formation and transport/deposition pathways, rapid testing procedures?
  Function: increase flow resistance, impact reactivity
  Fate: "on the wall" vs. "at the end of the channel"

Some Tools of the Trade: Side-stream Reactor Technology

- Operation-representative experimental setups (small samples/side stream technology) to screen technologies fast and identify input physicochemical parameters for modeling.

SAE 2000-01-1016, 2006-01-0874

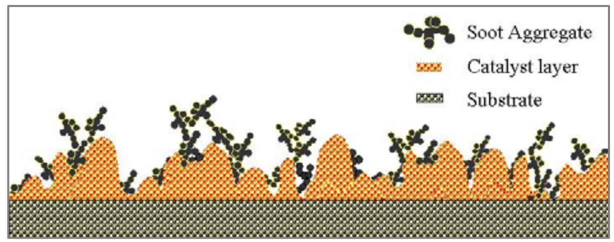


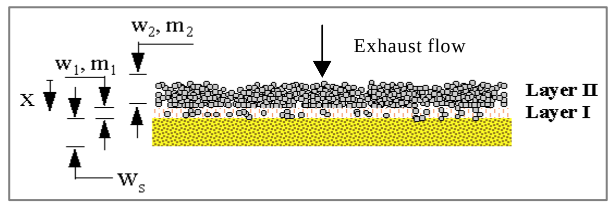


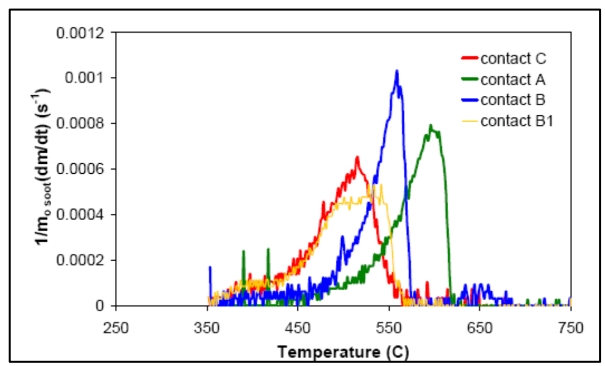
Some Tools of the Trade: "Coating"/Functionalization Technologies

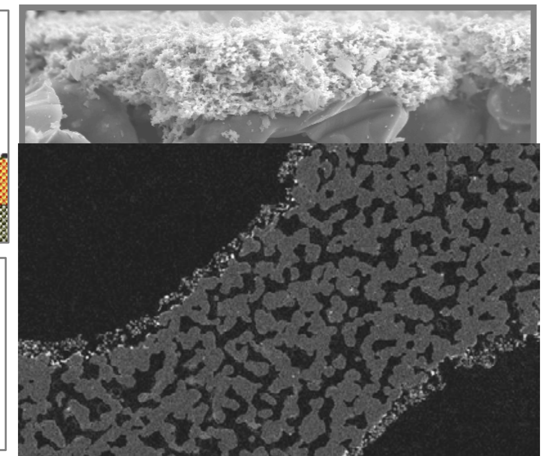
- DPF functionalization technologies for high filtration efficiency, low pressure drop and promotion of soot-catalyst contact for direct, sustained oxidation (2-Layer concept of catalytic coating).

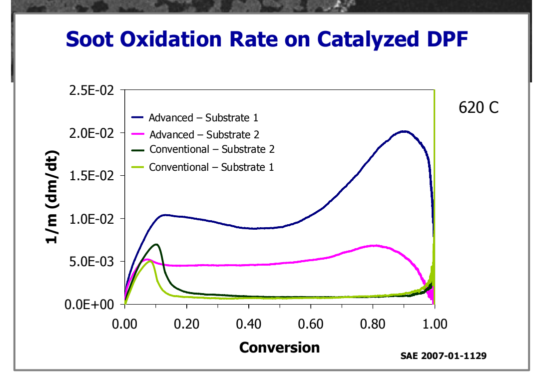
Konstandopoulos & Kostoglou (1998)
SAE 2005-01-0670, SAE 2008-01-0621





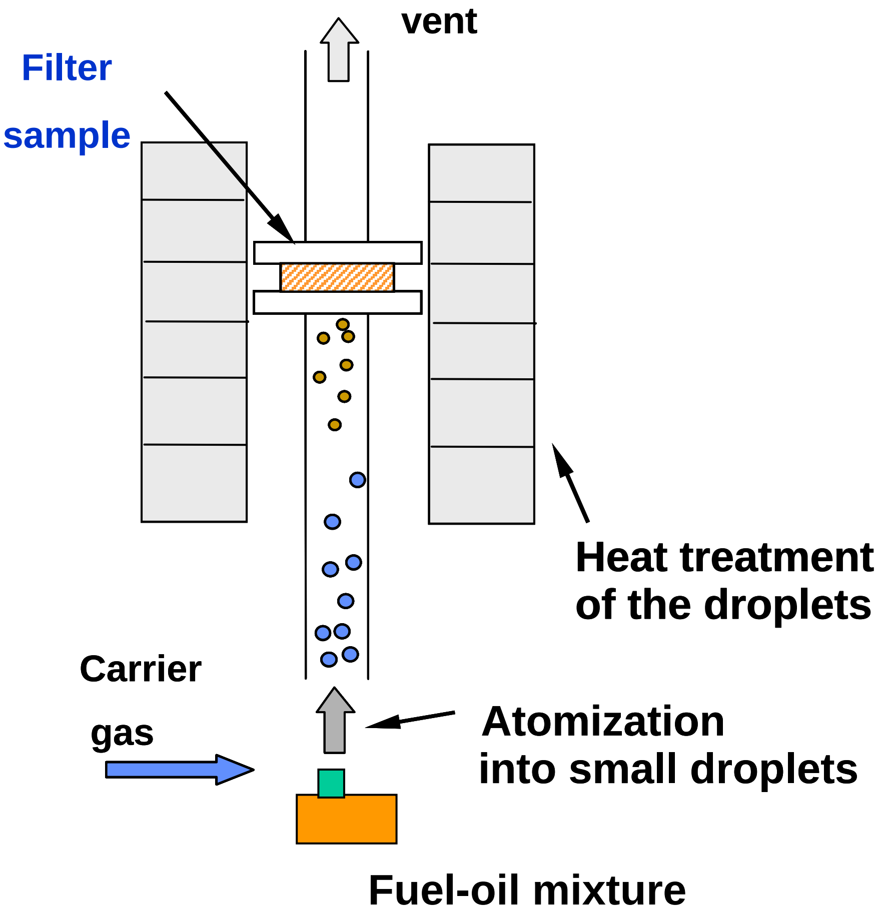




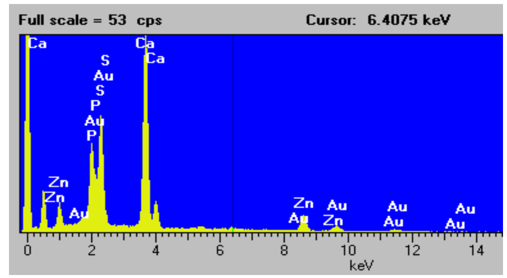


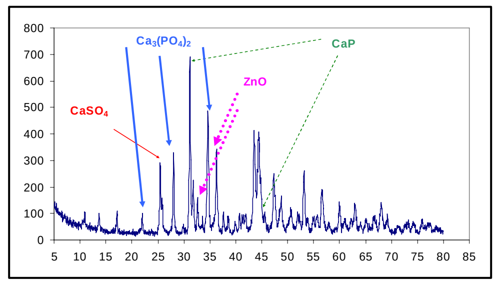
Some Tools of the Trade:
Rapid Ash/Aging Rig

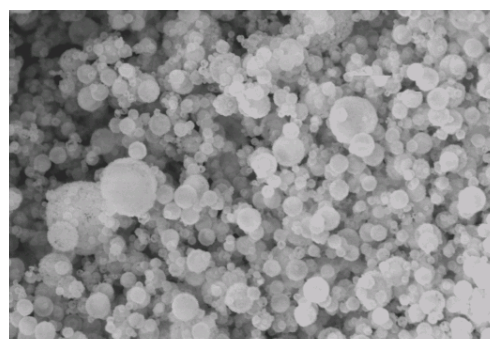
Fast ash aging deposition rig based on Aerosol Spray Pyrolysis of fuel-oil mixtures



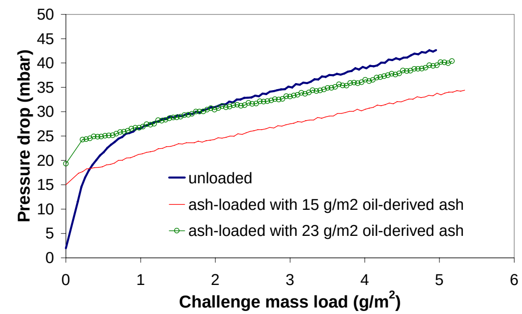
Ash Analytical Studies

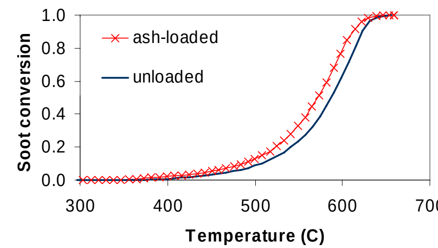




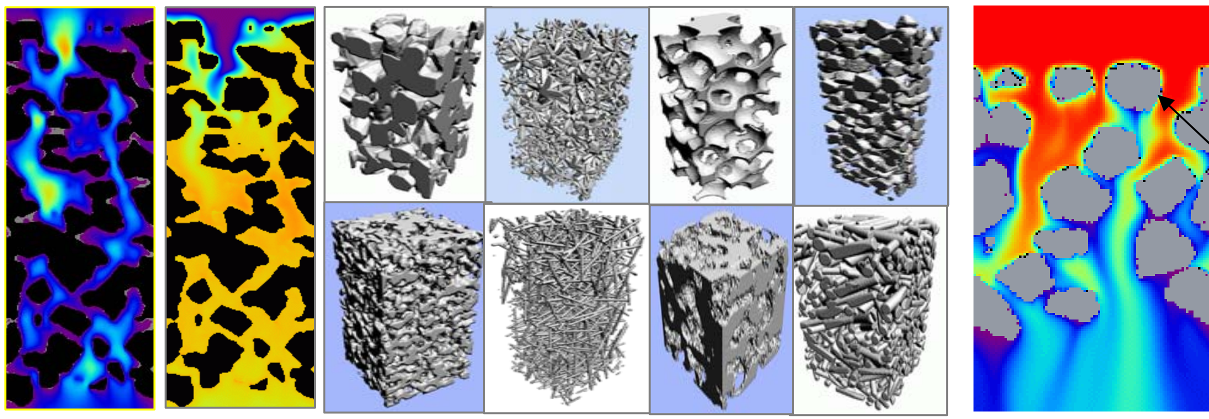


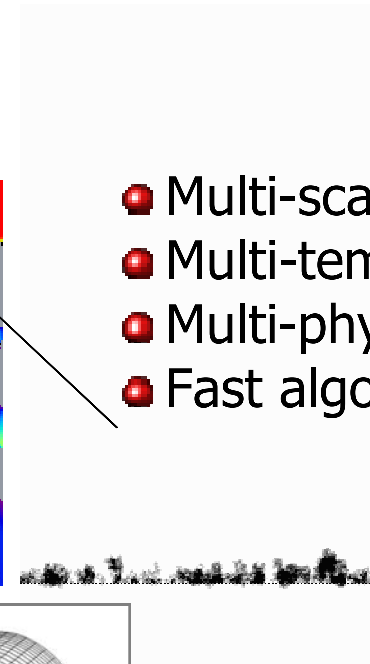
Engine Testing of Ash Loaded Samples



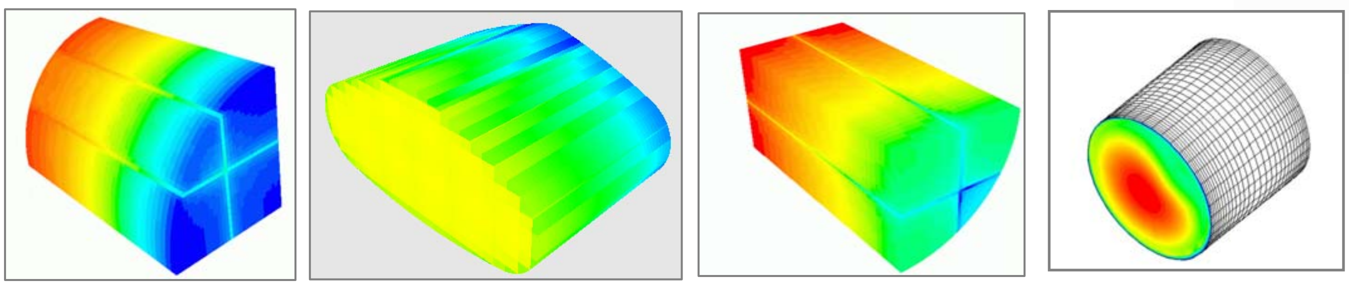


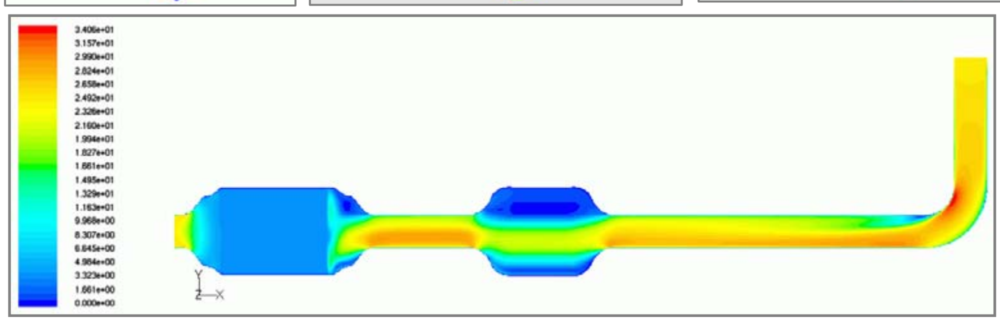
Some Tools of the Trade: Modeling & Simulation



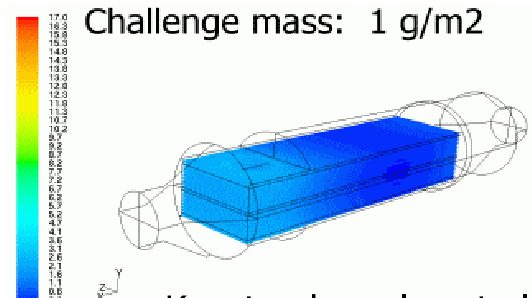


- Multi-scale
- Multi-temporal
- Multi-physicochemical
- Fast algorithms



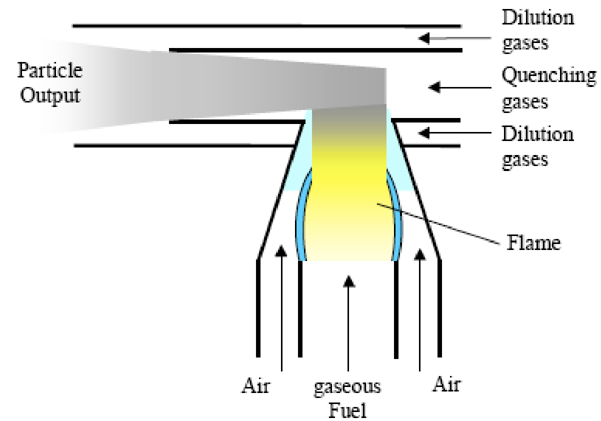


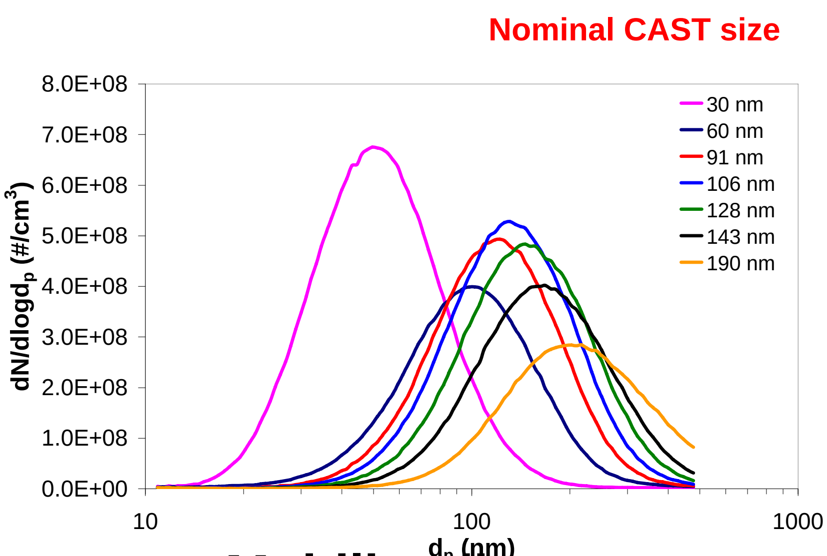
Challenge mass: 1 g/m2



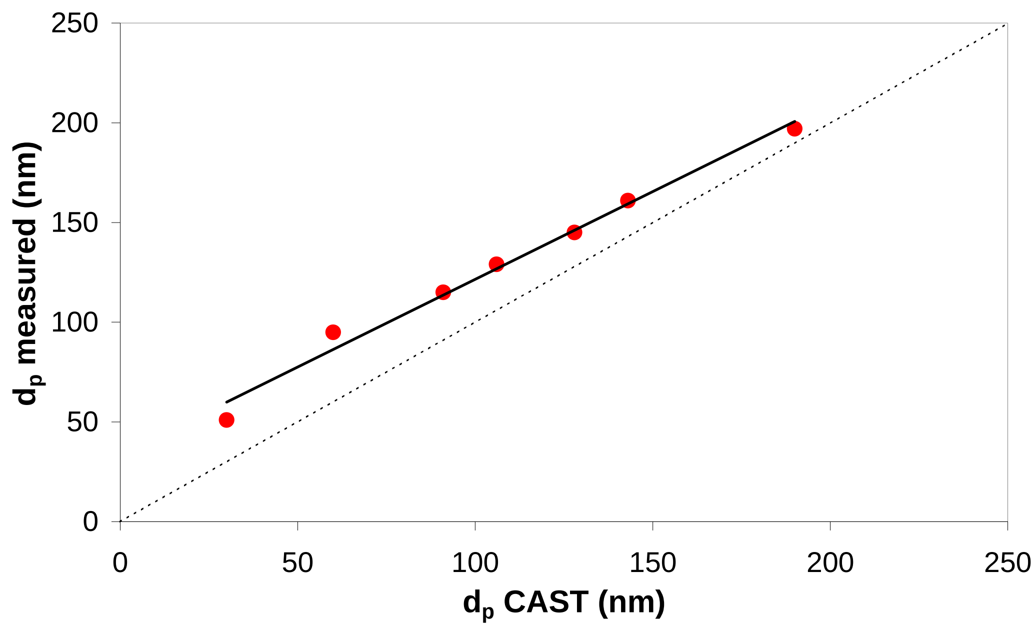
Konstandopoulos et al. (1989-2009)

Combustion Aerosol Standard (CAST) Burner

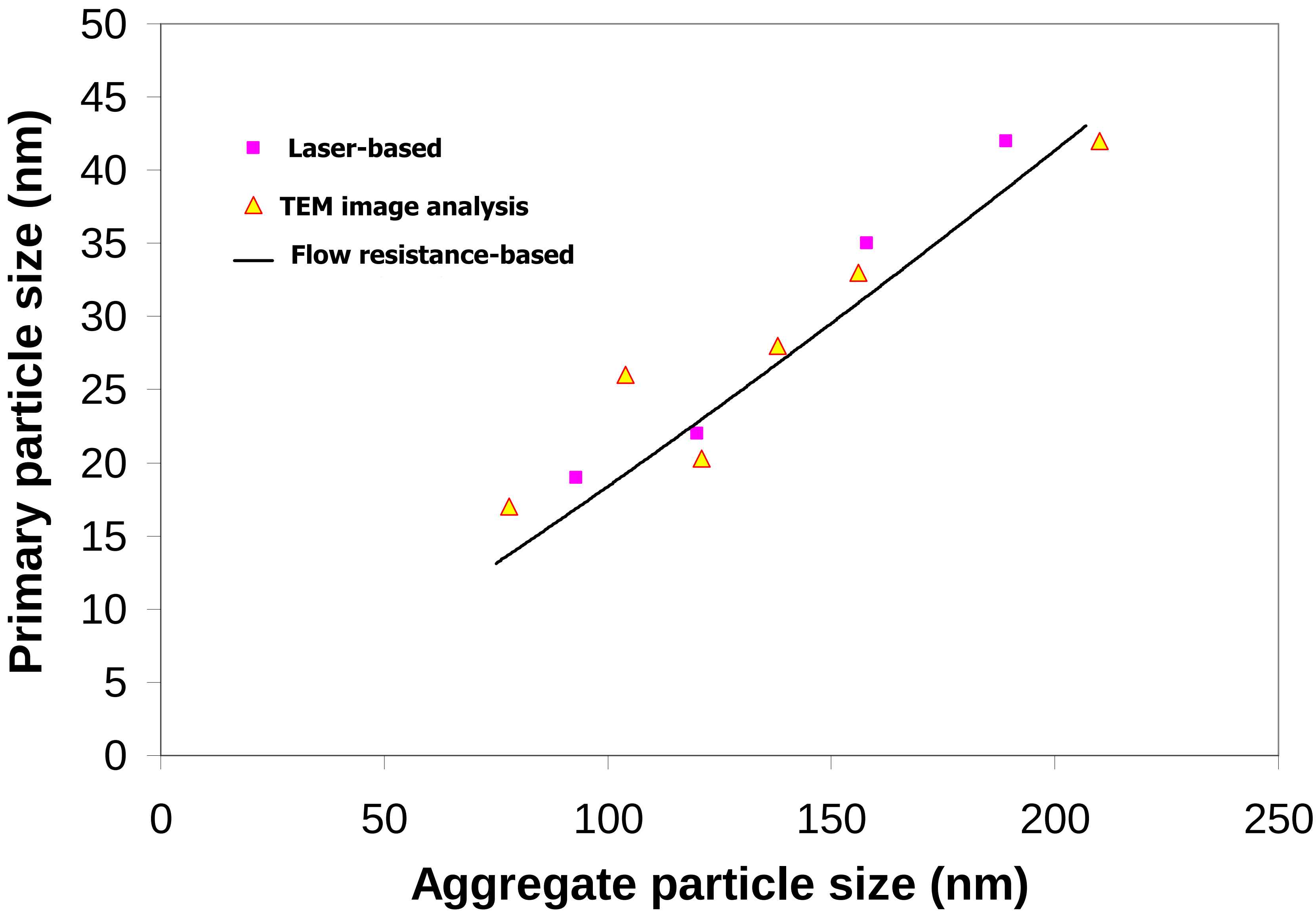




Mobility diameter

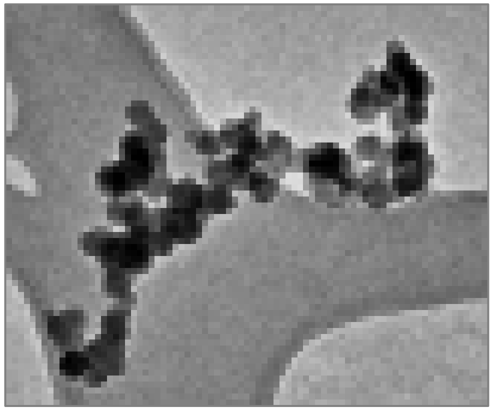


CAST Primary Particle vs. Mobility Diameter



Diesel Soot Aggregate Morphology

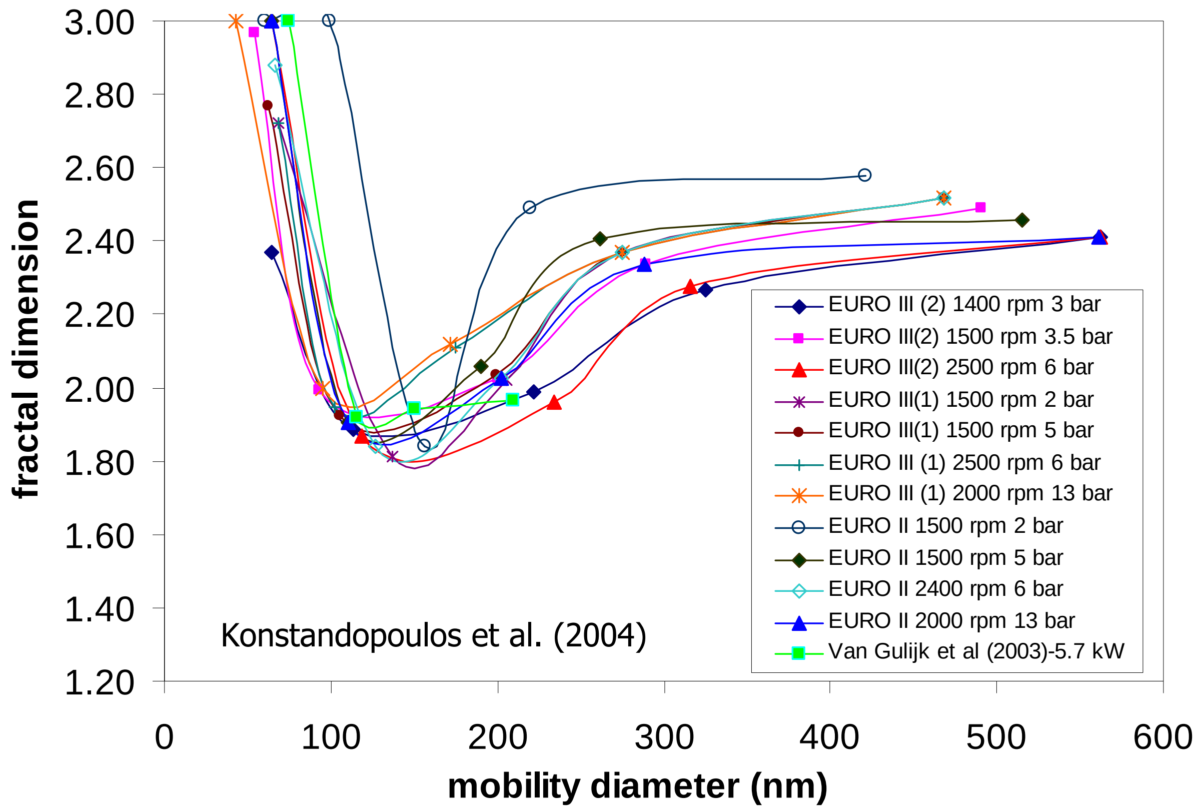
Soot fractal aggregate



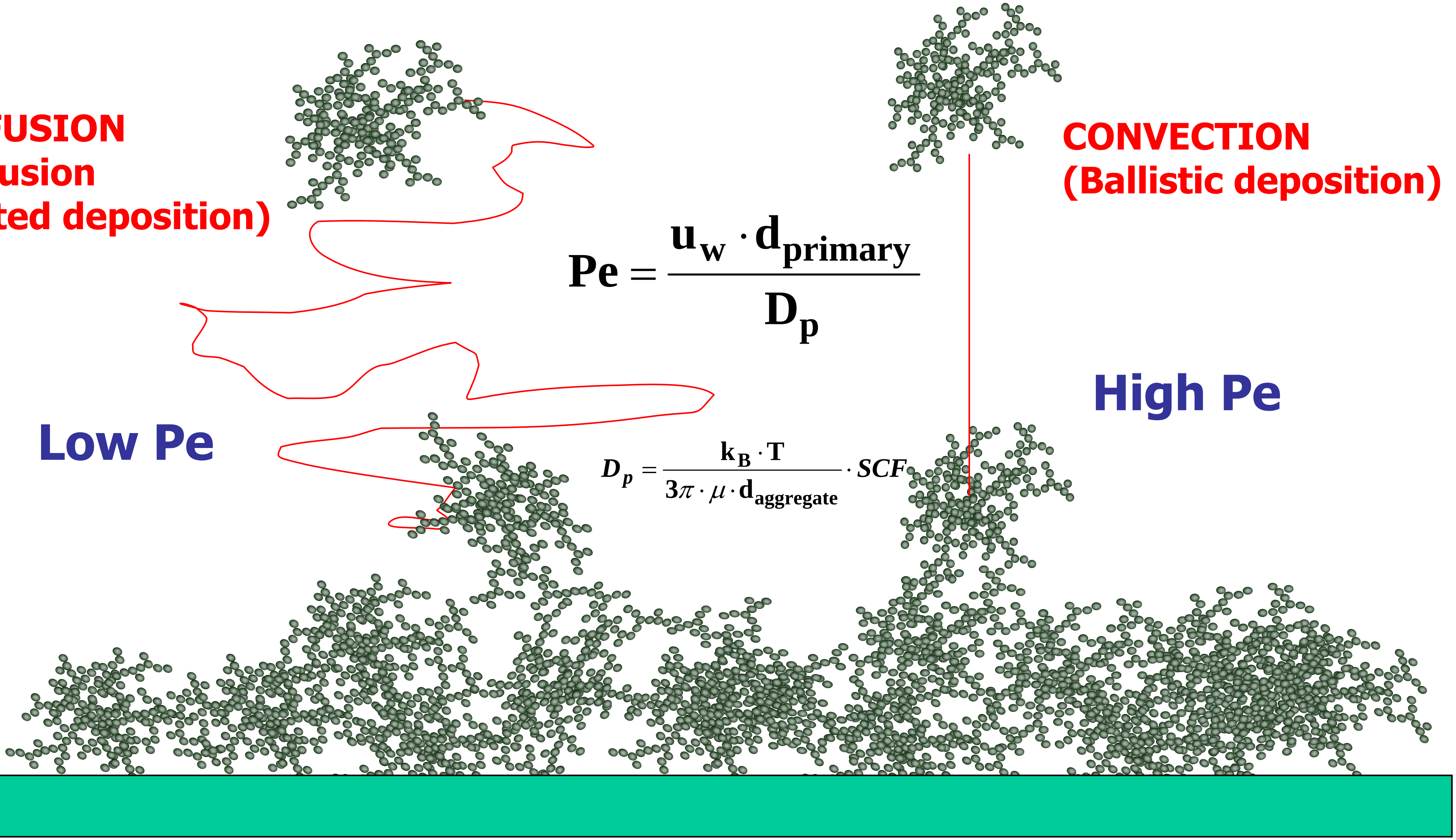
Number of primary particles per aggregate

$$N_A = k_g \left[\frac{D_g}{d_0} \right]^{D_f}$$

3 different diesel engines & 1 gen set



Soot Cake Formation Mechanism

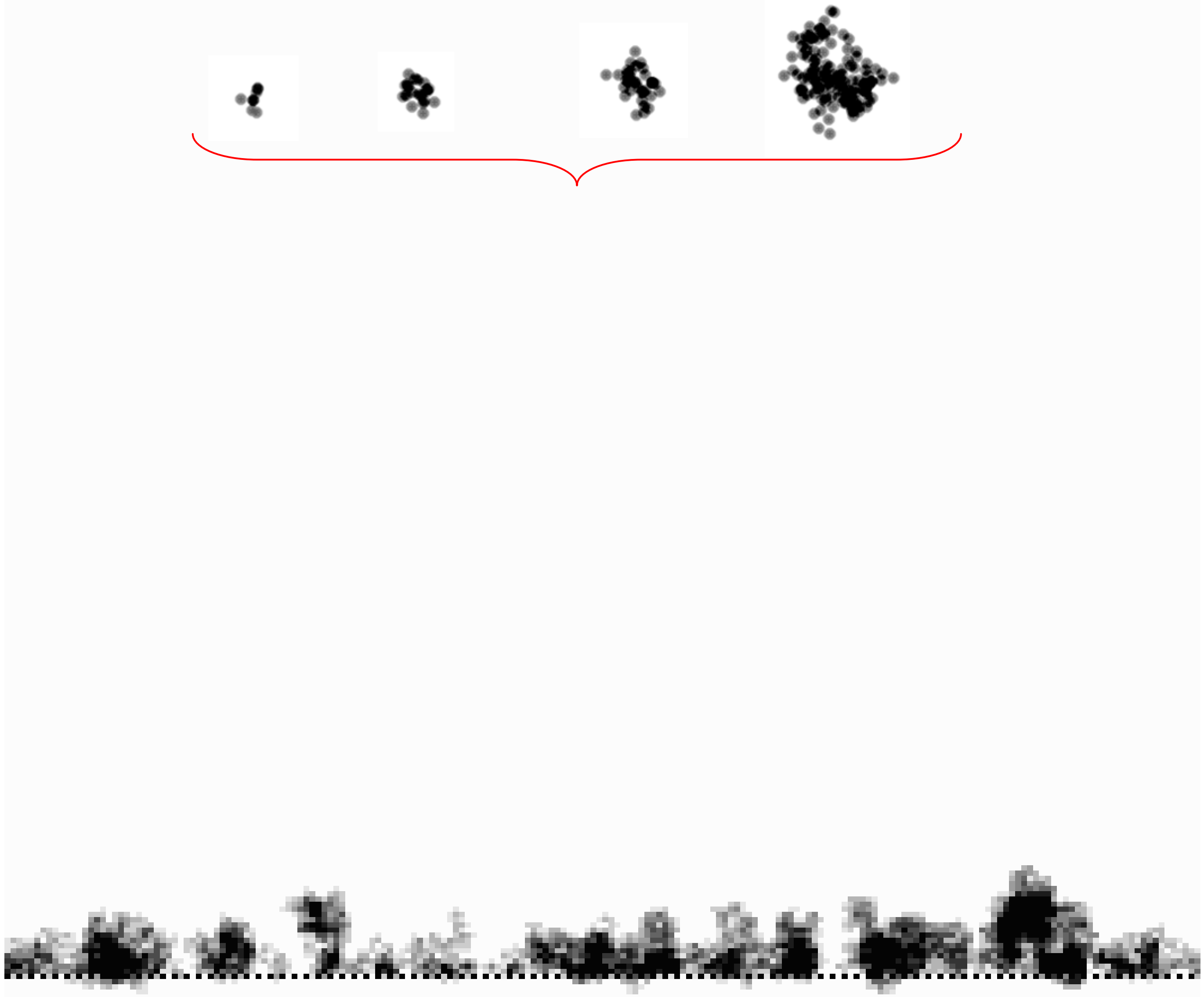


$$\mathbf{Pe} = \frac{\mathbf{u_w} \cdot \mathbf{d_{primary}}}{\mathbf{D_p}}$$

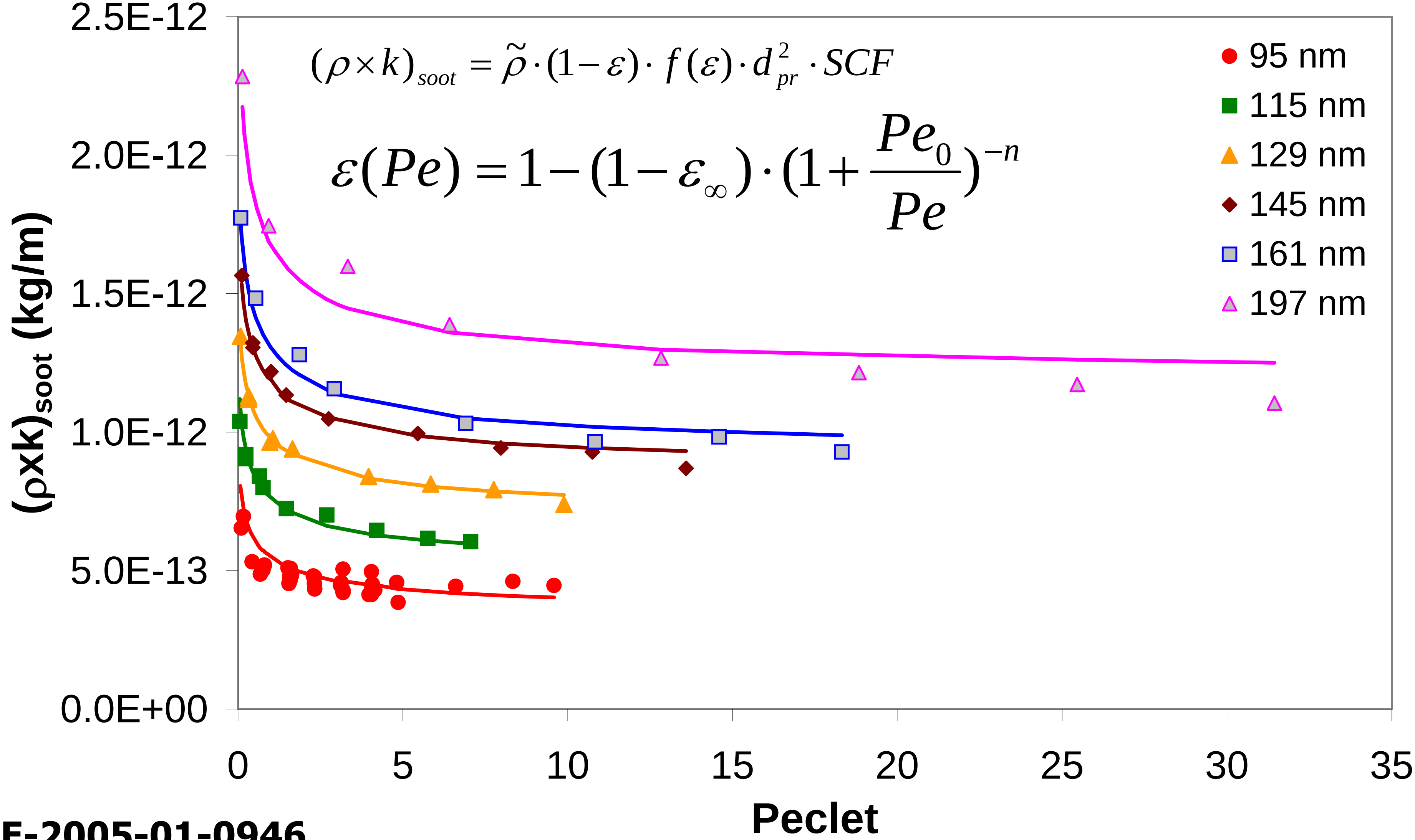
$$D_p = \frac{\mathbf{k_B} \cdot \mathbf{T}}{3\pi \cdot \mu \cdot \mathbf{d_{aggregate}}} \cdot SCF$$

Konstandopoulos et al. (2002, 2005)

Soot aggregate deposit formation

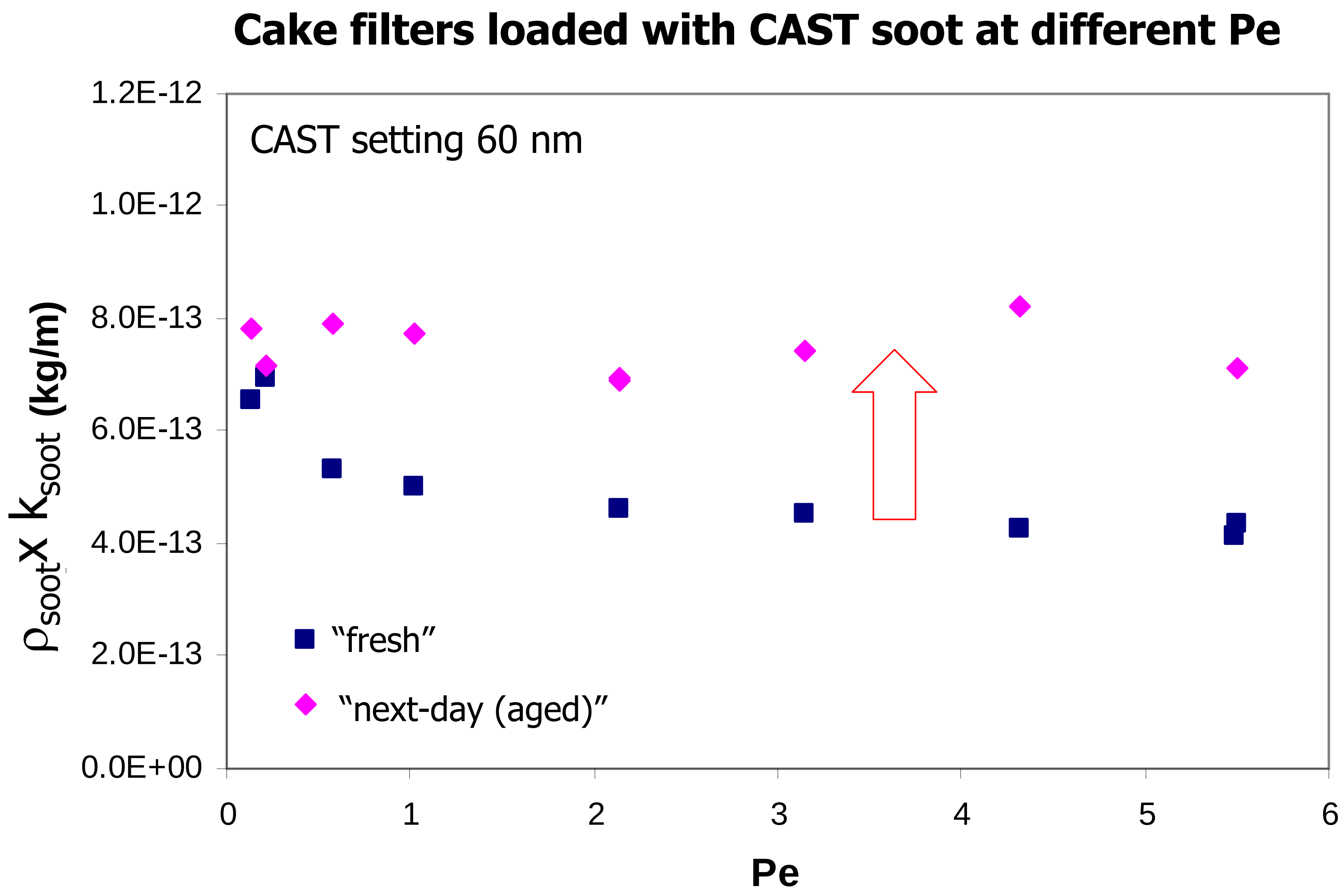


Soot flow resistance $(\rho_{soot} x\ k_{soot})^{-1}$



SAE-2005-01-0946

"Next day" increase of flow conductance (ρ_{soot}x k_{soot})



What causes the increase of flow conductance (ρ_{soot}x k_{soot})?

$$(\rho_{soot} \times k_{soot}) = \tilde{\rho} \cdot (1-\varepsilon) \cdot f(\varepsilon) \cdot d_{pr}^2 \cdot SCF(d_{pr}, T)$$

If d_{pr} is increased by condensation of ambient humidity then ε is reduced

If capillary condensation menisci induce homogeneous compaction then ε is reduced

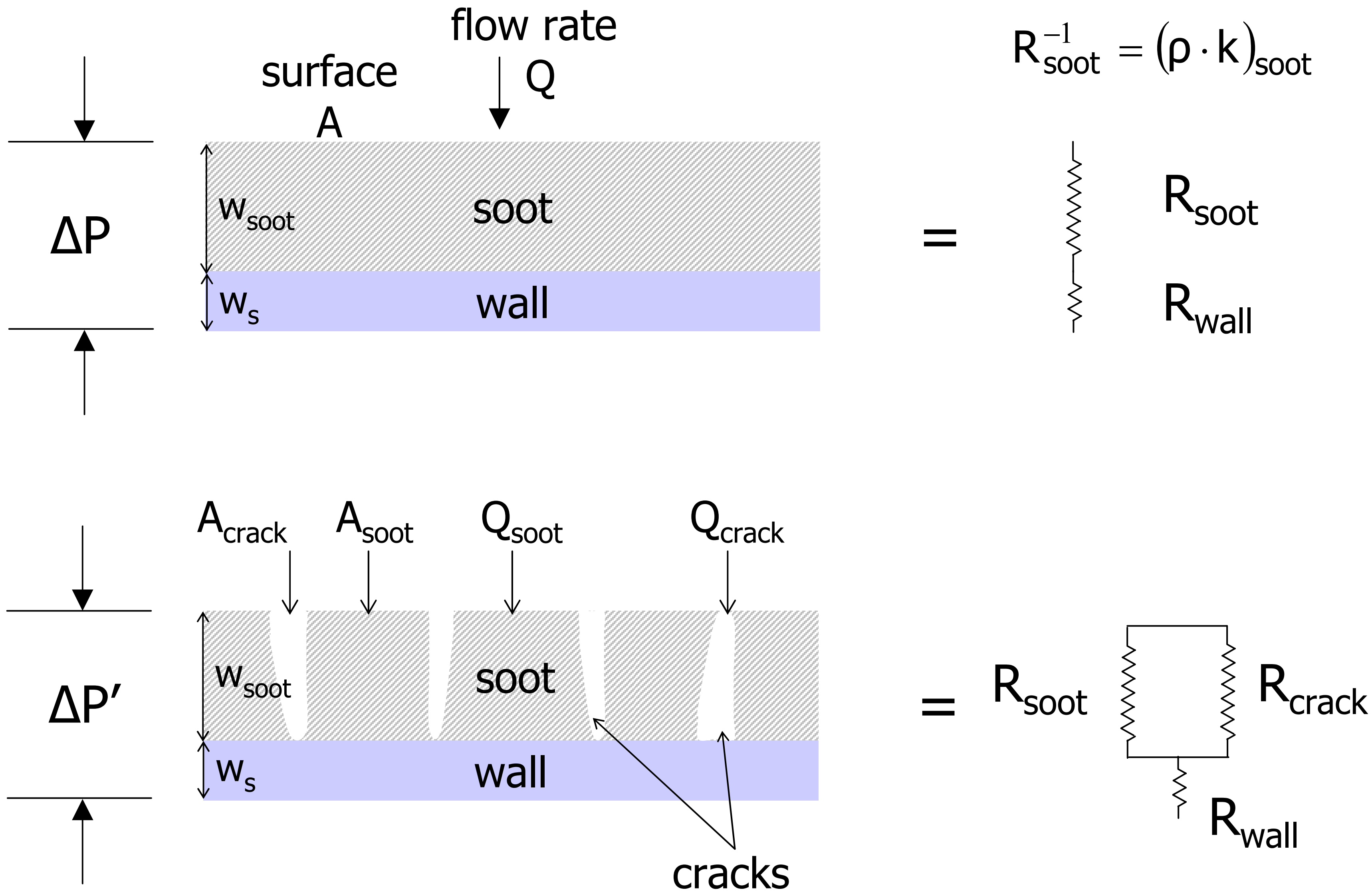
} ρ_{soot}x k_{soot} is reduced

A non-homogeneous restructuring must occur similar to:

Crack images from google

Mechanism of increase of flow conductance ($\rho_{soot} \times k_{soot}$)

$$R_{soot}^{-1} = (\rho \cdot k)_{soot}$$



Mathematical Model of Cracked Soot Deposit

$$\Delta P = \frac{\mu}{k_{clean}} \cdot \frac{Q}{A} \cdot w_s + \frac{\mu}{(\rho \cdot k)_{soot}} \cdot \frac{Q}{A^2} \cdot m_{soot} \quad \textbf{(1)}$$

$$\Delta P' = \frac{\mu}{k_{clean}} \cdot \frac{q_{soot}}{A_{soot}} \cdot w_s + \frac{\mu}{(\rho \cdot k)_{soot}} \cdot \frac{q_{soot}}{A_{soot}^{\ 2}} \cdot m_{soot} \quad \textbf{(2)}$$

$$\Delta P' = \frac{\mu}{k_{clean}} \cdot \frac{q_{crack}}{A_{crack}} \cdot w_s \quad \textbf{(3)}$$

$$q_{soot} + q_{crack} = Q' \Rightarrow \begin{cases} q_{soot} = (1 - \varphi) \cdot Q' \\ q_{crack} = \varphi \cdot Q' \end{cases} \quad \textbf{(4)}$$

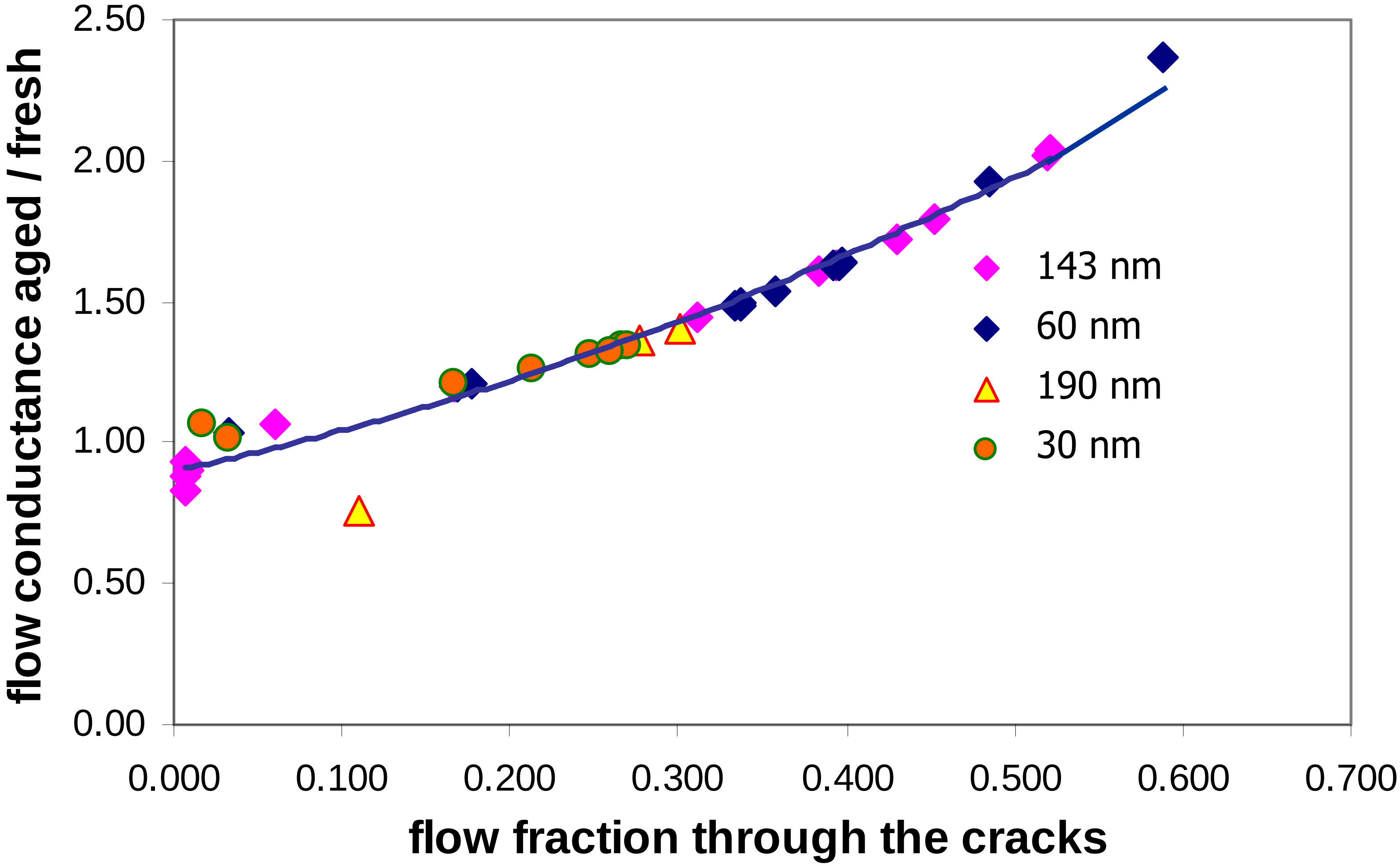
Flow Fraction through the cracks

$$A_{soot} + A_{crack} = A \Rightarrow \begin{cases} A_{soot} = (1 - \psi) \cdot A \\ A_{crack} = \psi \cdot A \end{cases} \quad \textbf{(5)}$$

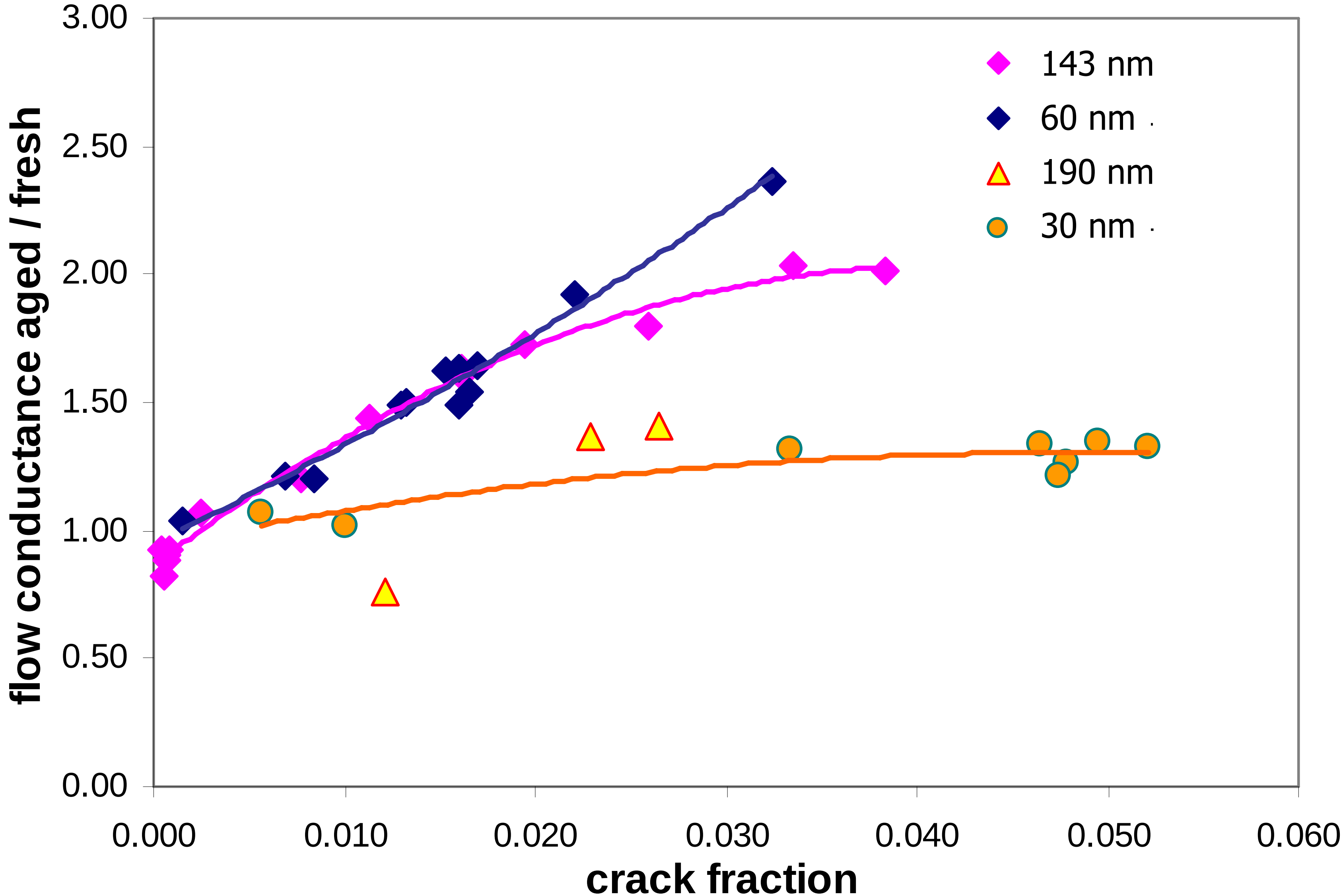
Crack Fraction

solve for φ and ψ

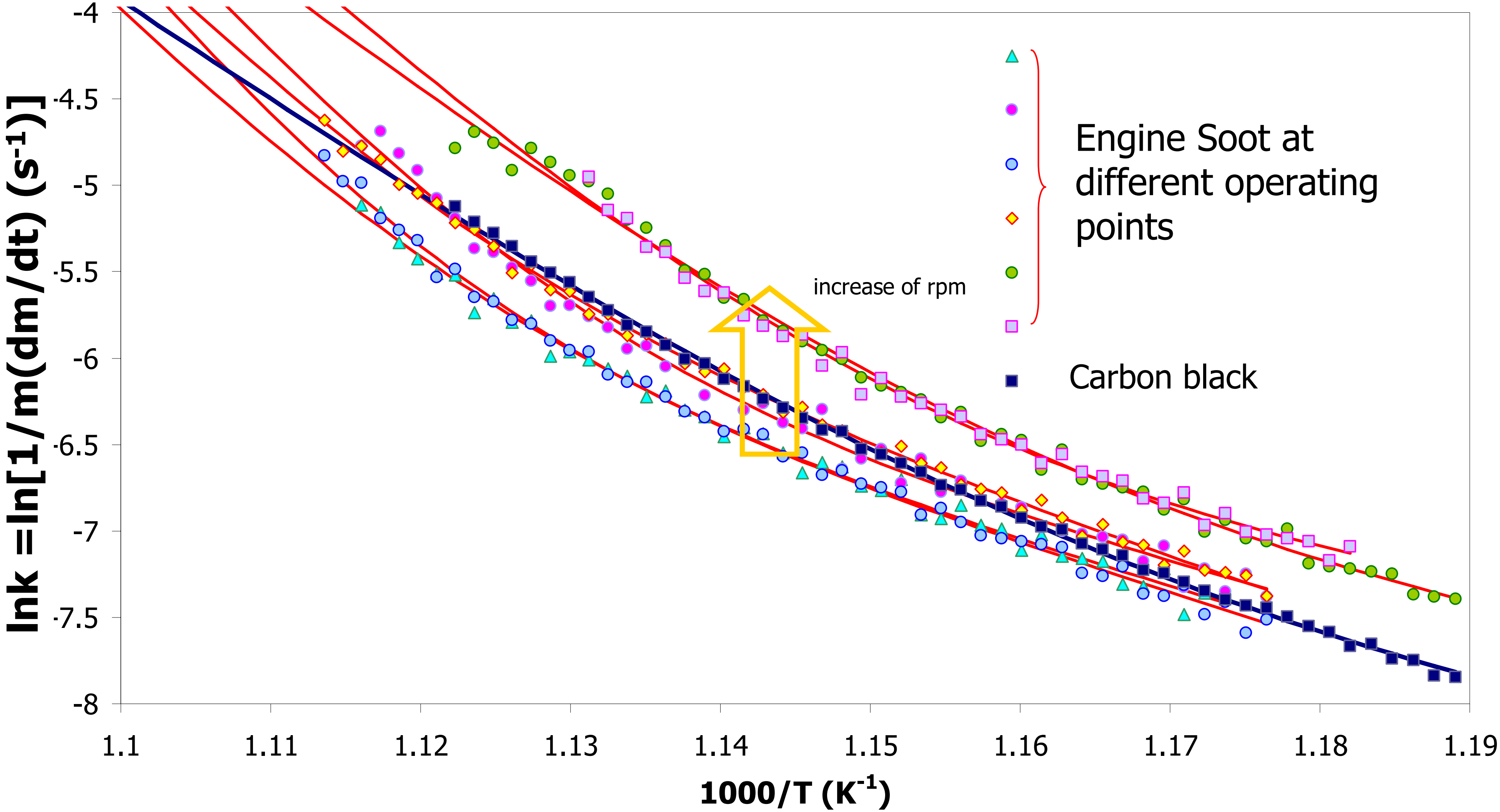
Increase of flow conductance (ρ_{soot} x k_{soot}) with flow fraction ϕ through the cracks



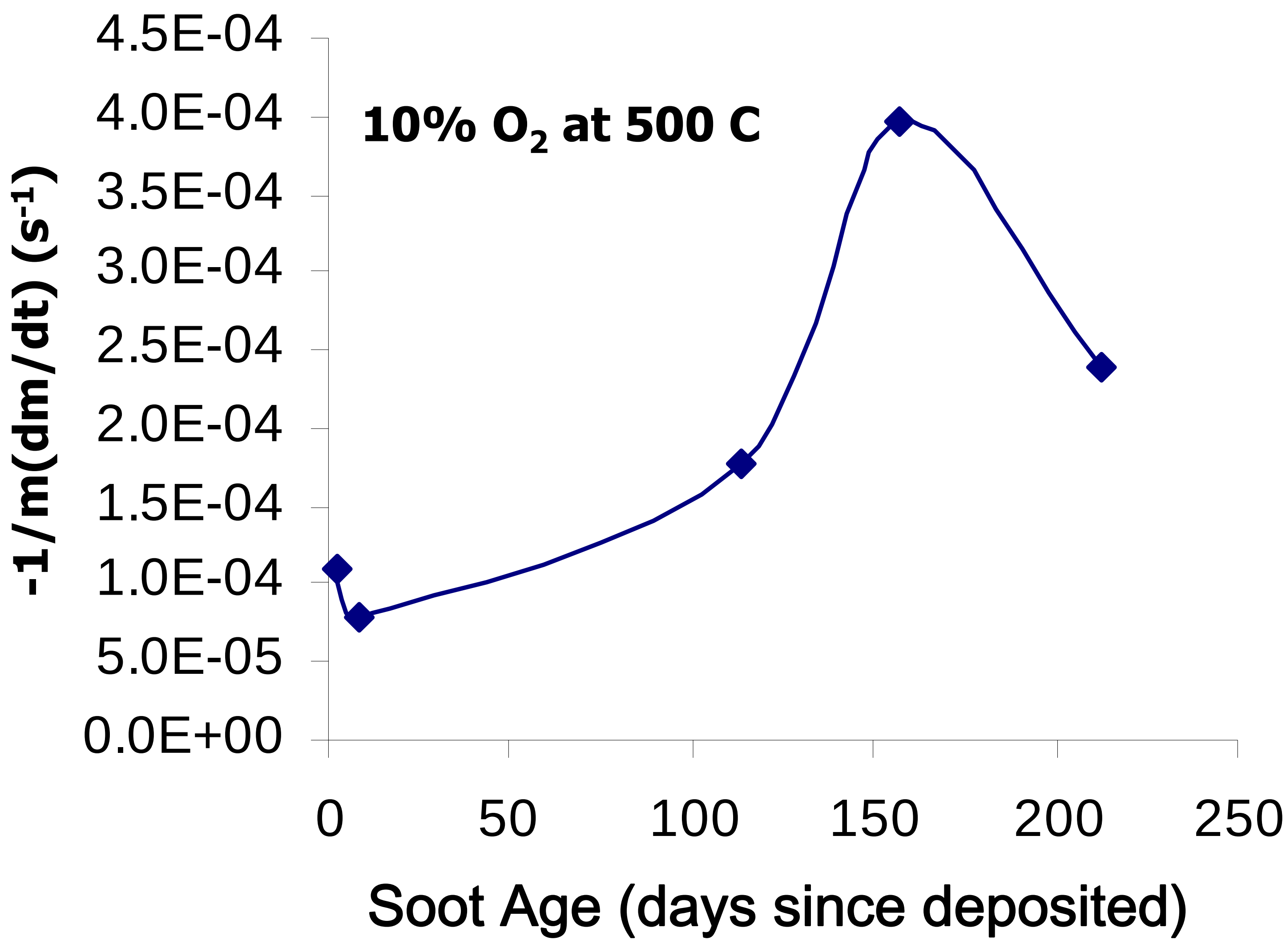
Increase of flow conductance (ρ_{soot} x k_{soot}) with crack fraction ψ



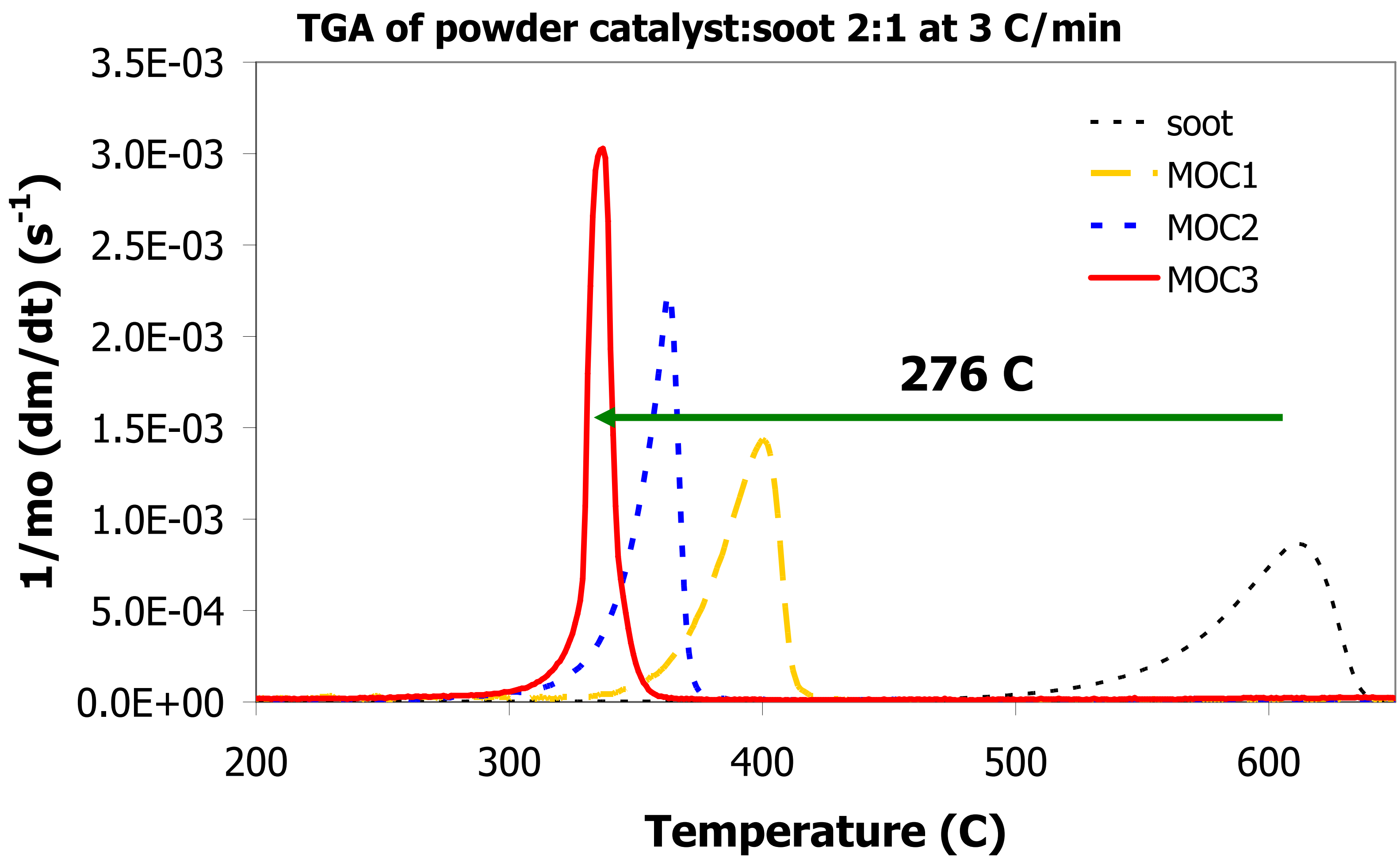
Variations in soot reactivity



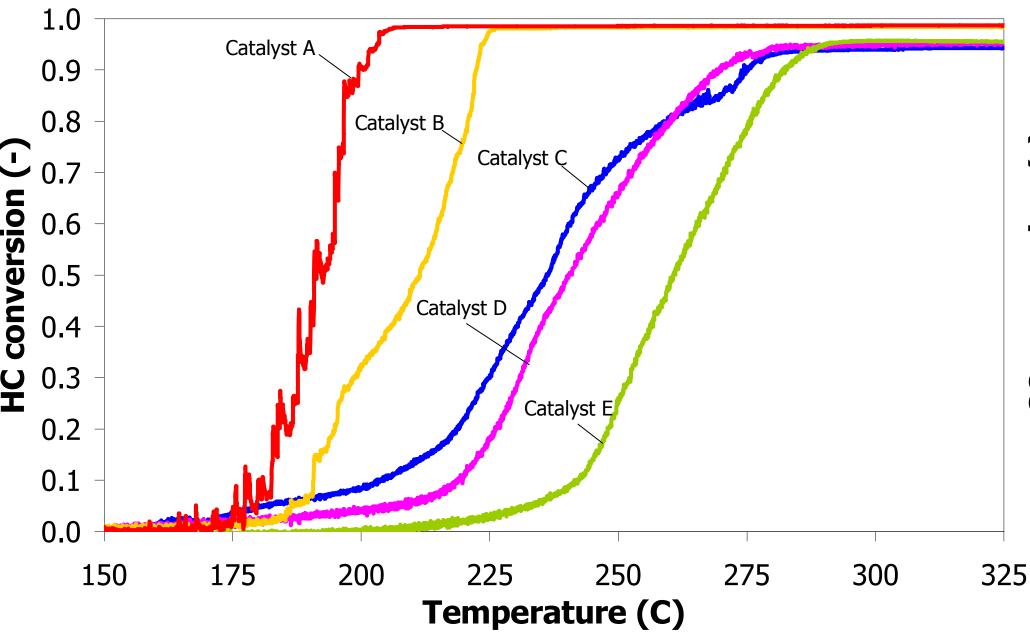
Effect of Aging on Soot Reactivity

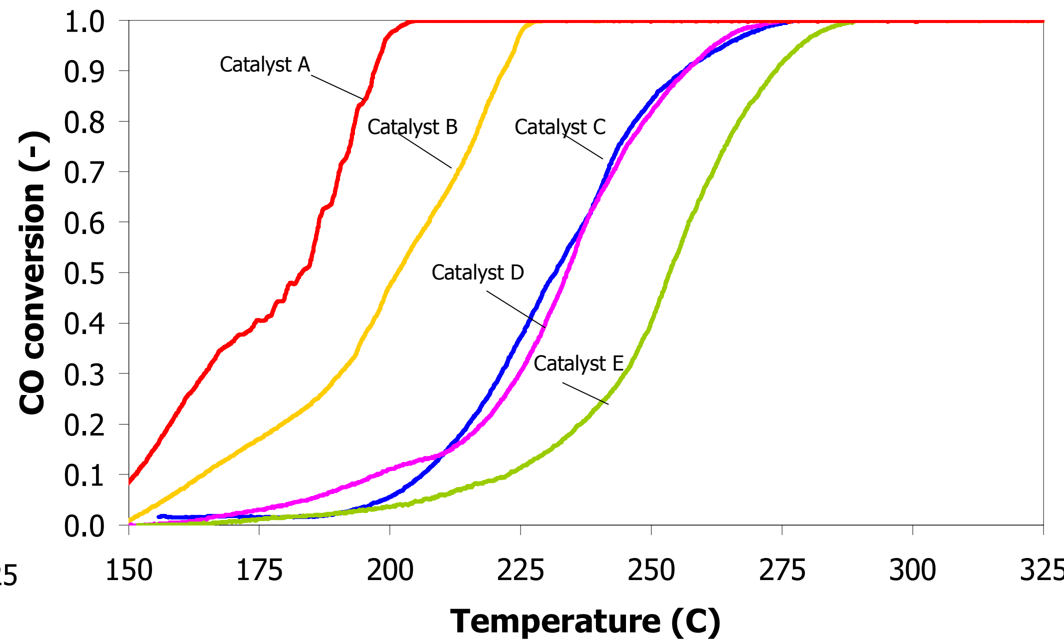


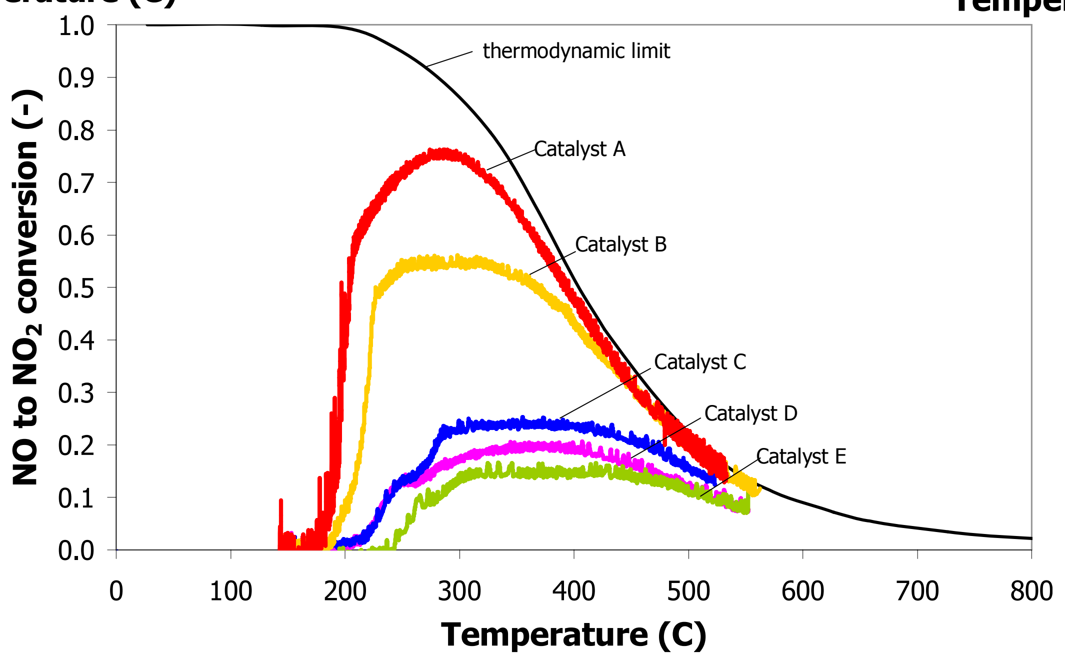
Soot Oxidation Catalyst Chemistry Development



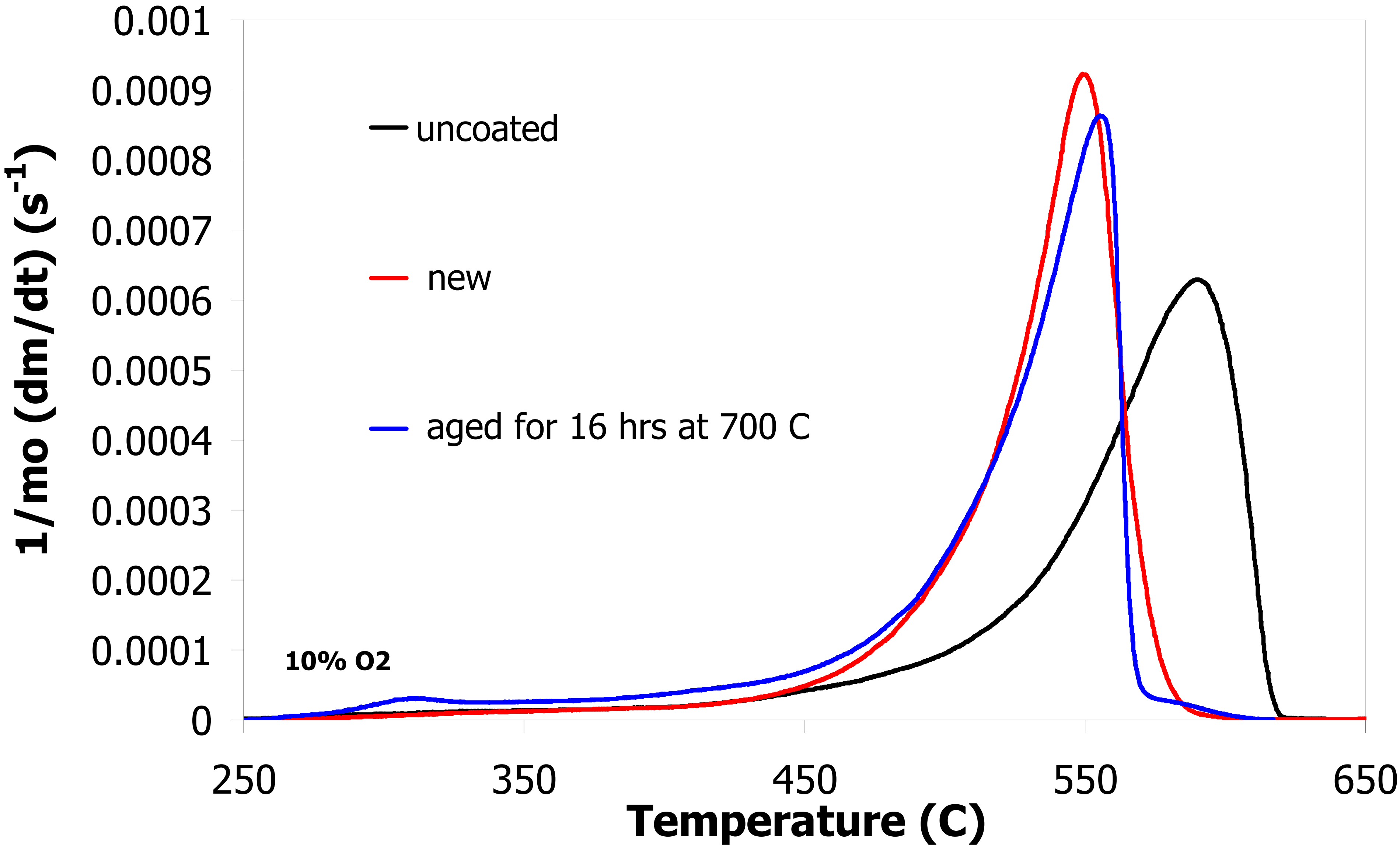
CO, HC and NO oxidation functions



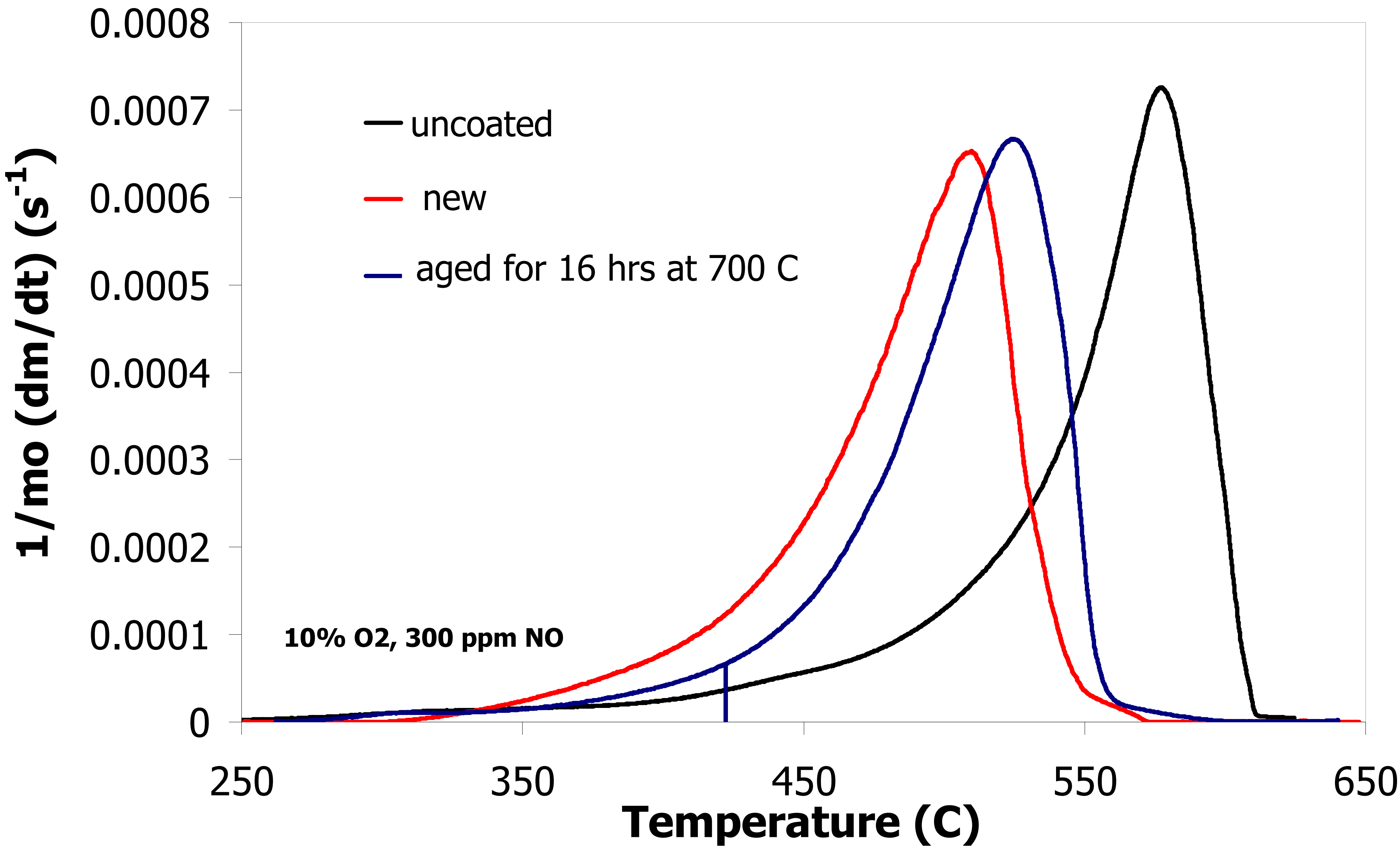




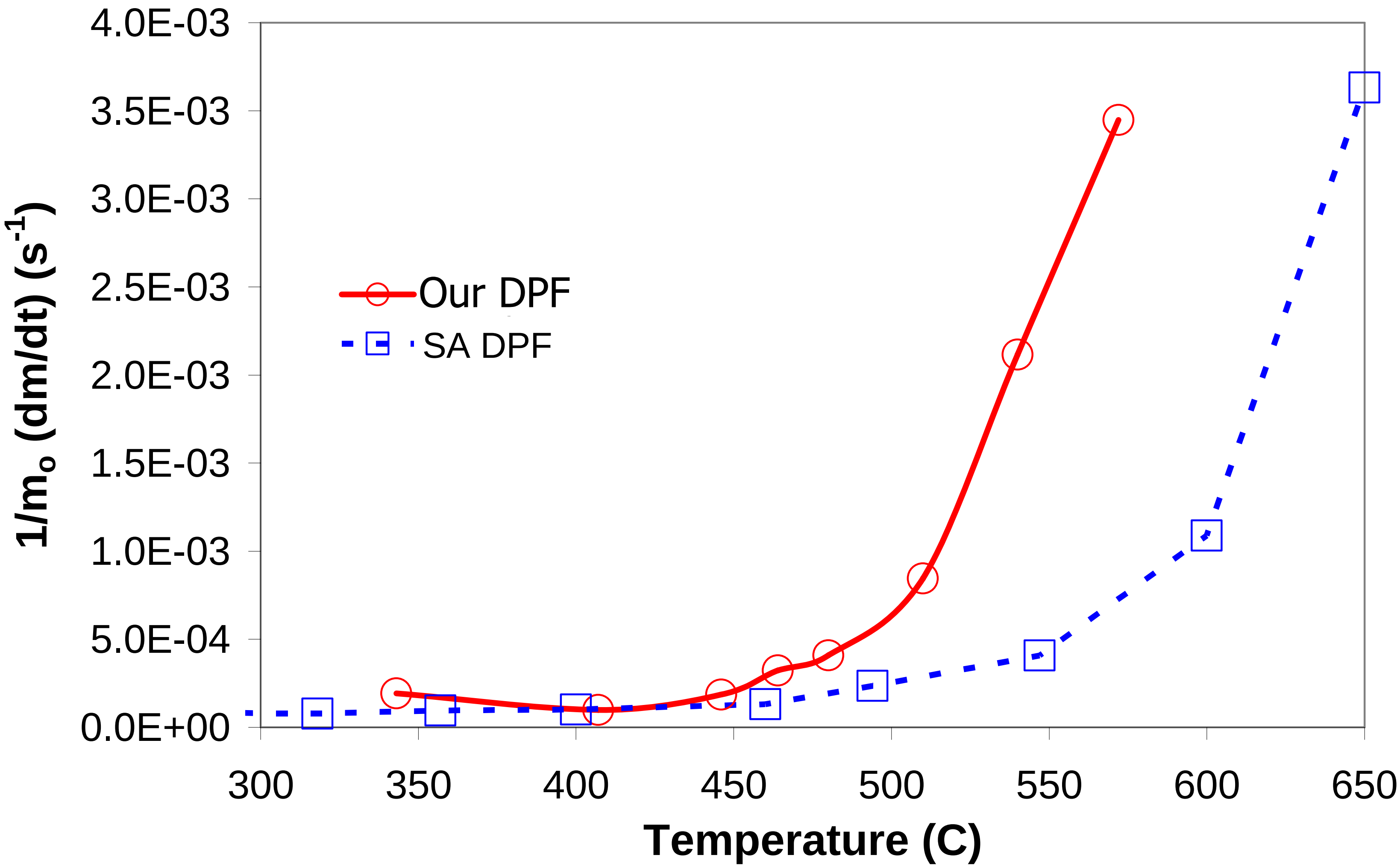
Effect of Thermal Aging on Direct Soot Reactivity



Effect of Thermal Aging on Indirect Soot Reactivity



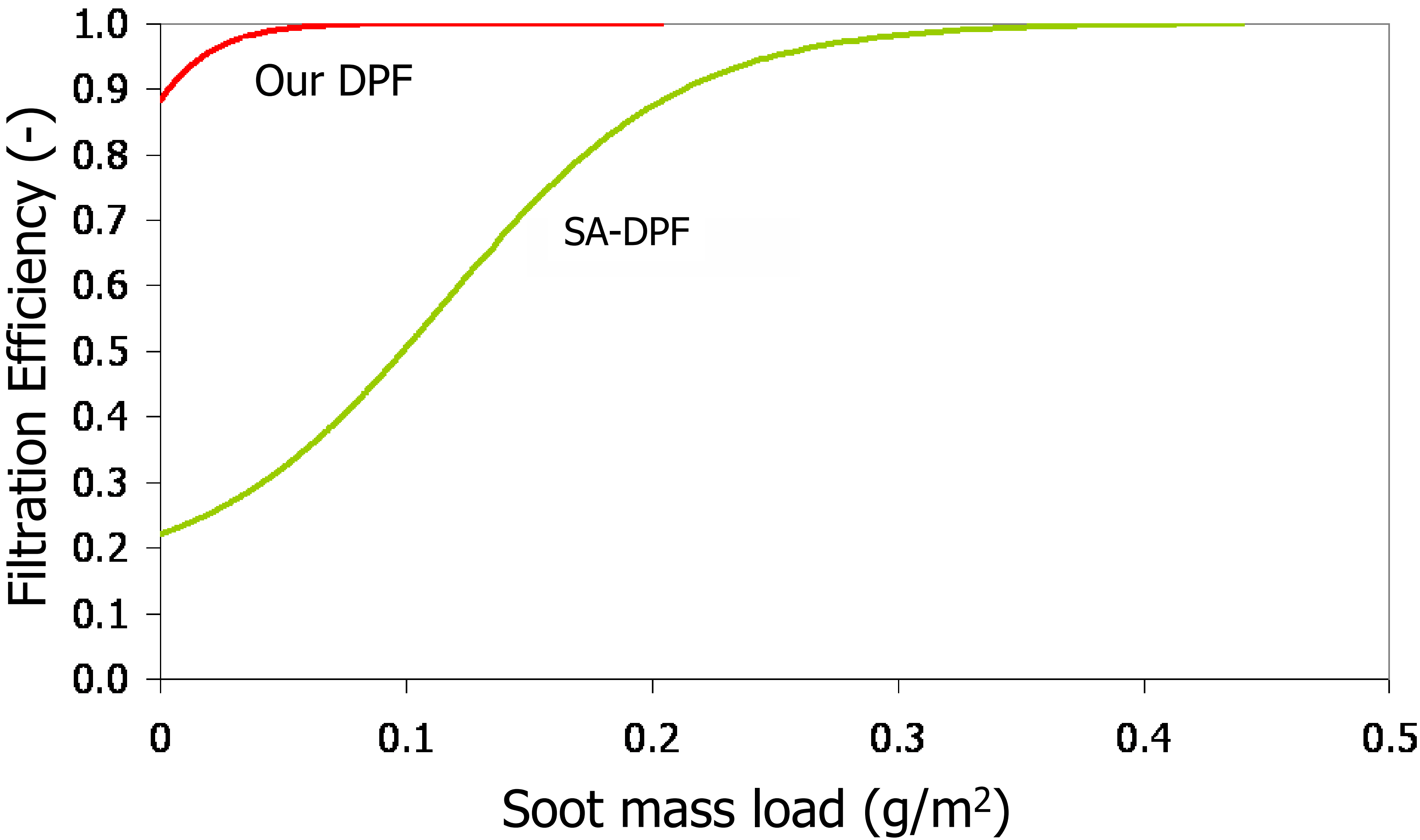
Scaled Up DPF on Engine Bench



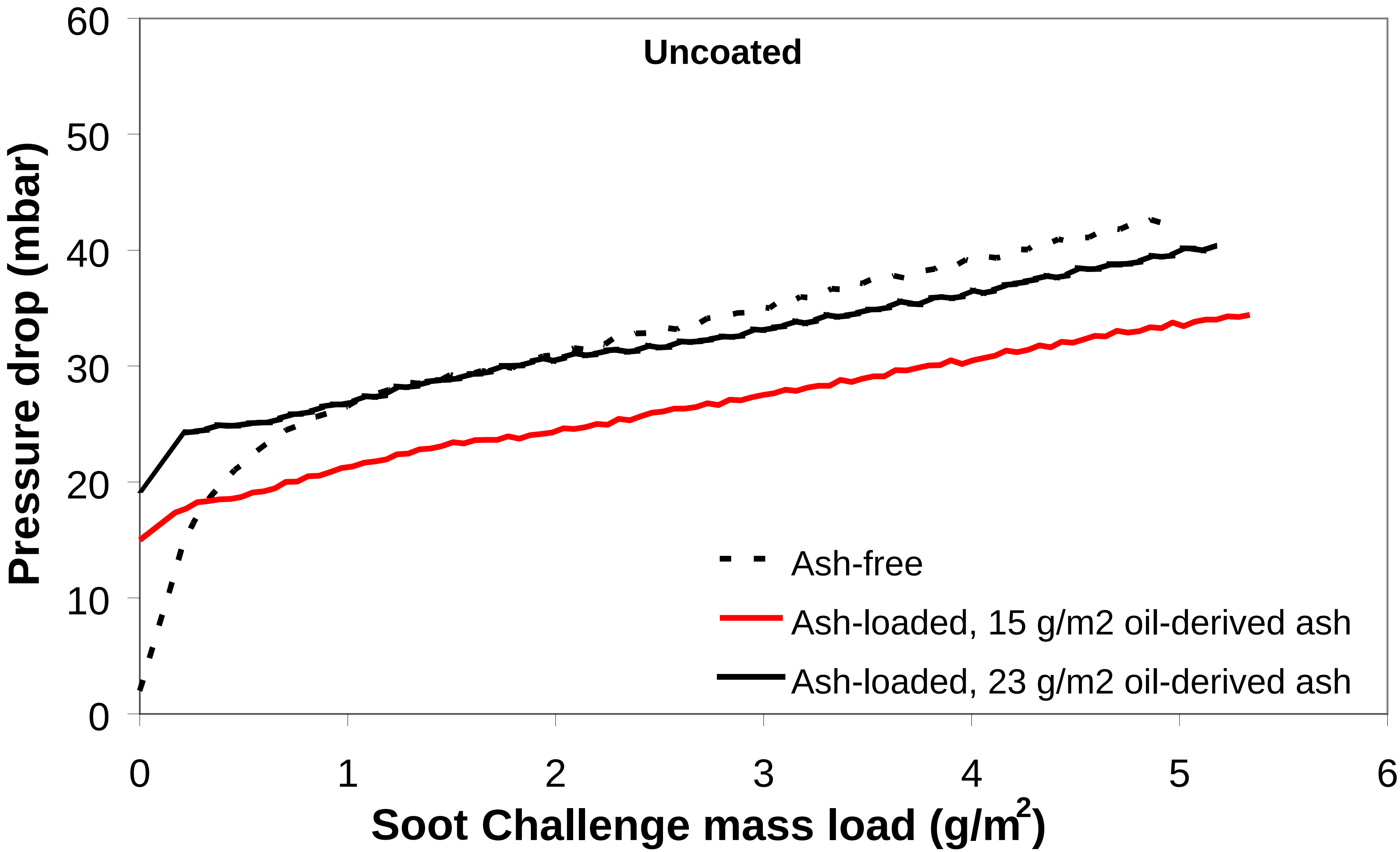
4 times higher soot oxidation rate at 550 C compared to the SA DPF.

SAE 2009-01-0287

Filtration Efficiency by Number

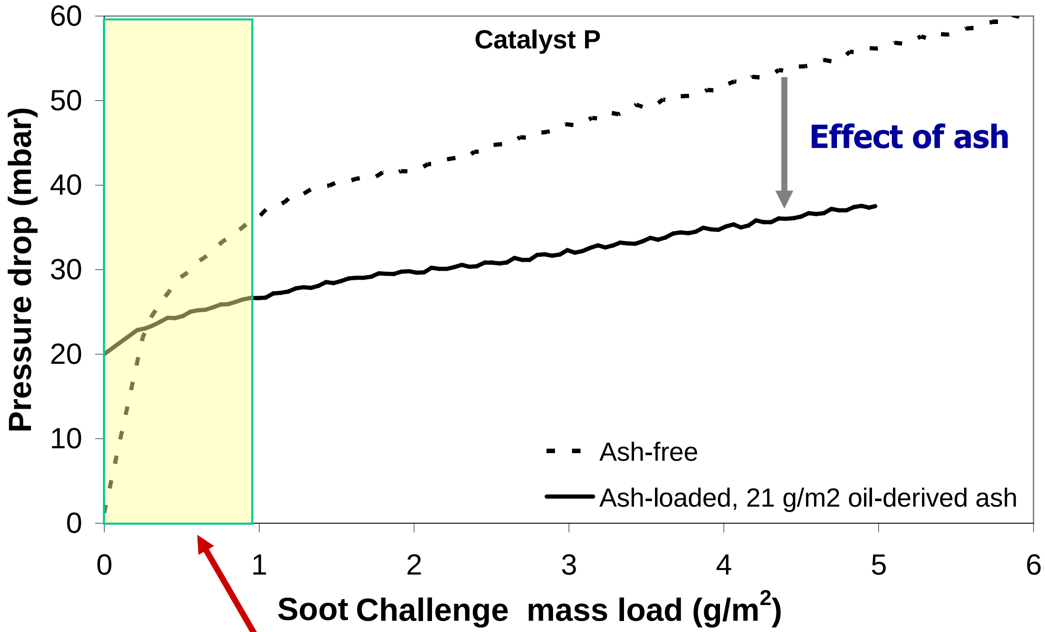


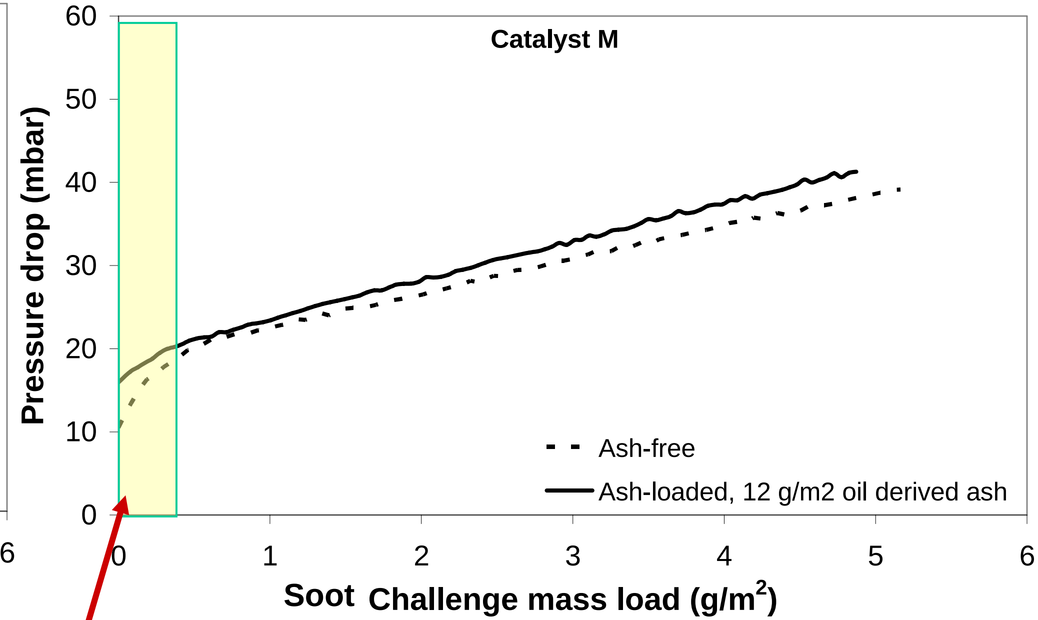
Soot Loading – Effect of Ash on Uncoated Filters



Soot Loading – Effect of Ash on Catalyzed Filters

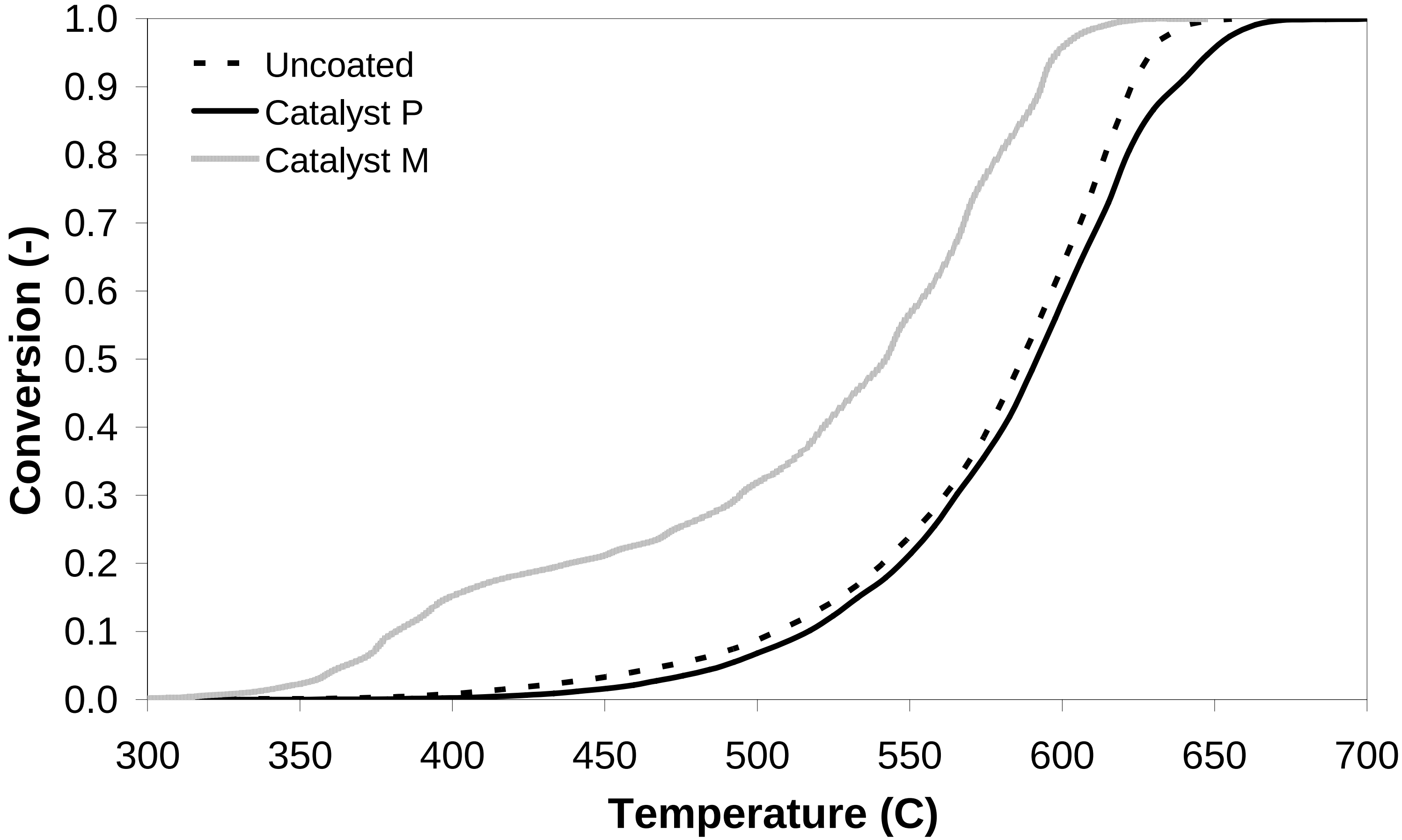
Catalyzed filter with Catalyst P and M by conventional wet chemistry methods





The existence and extent of the deep bed filtration regime (irrespective of whether the filter is coated or uncoated) determines how the accumulating ash layer impacts the filter pressure drop during soot loading.

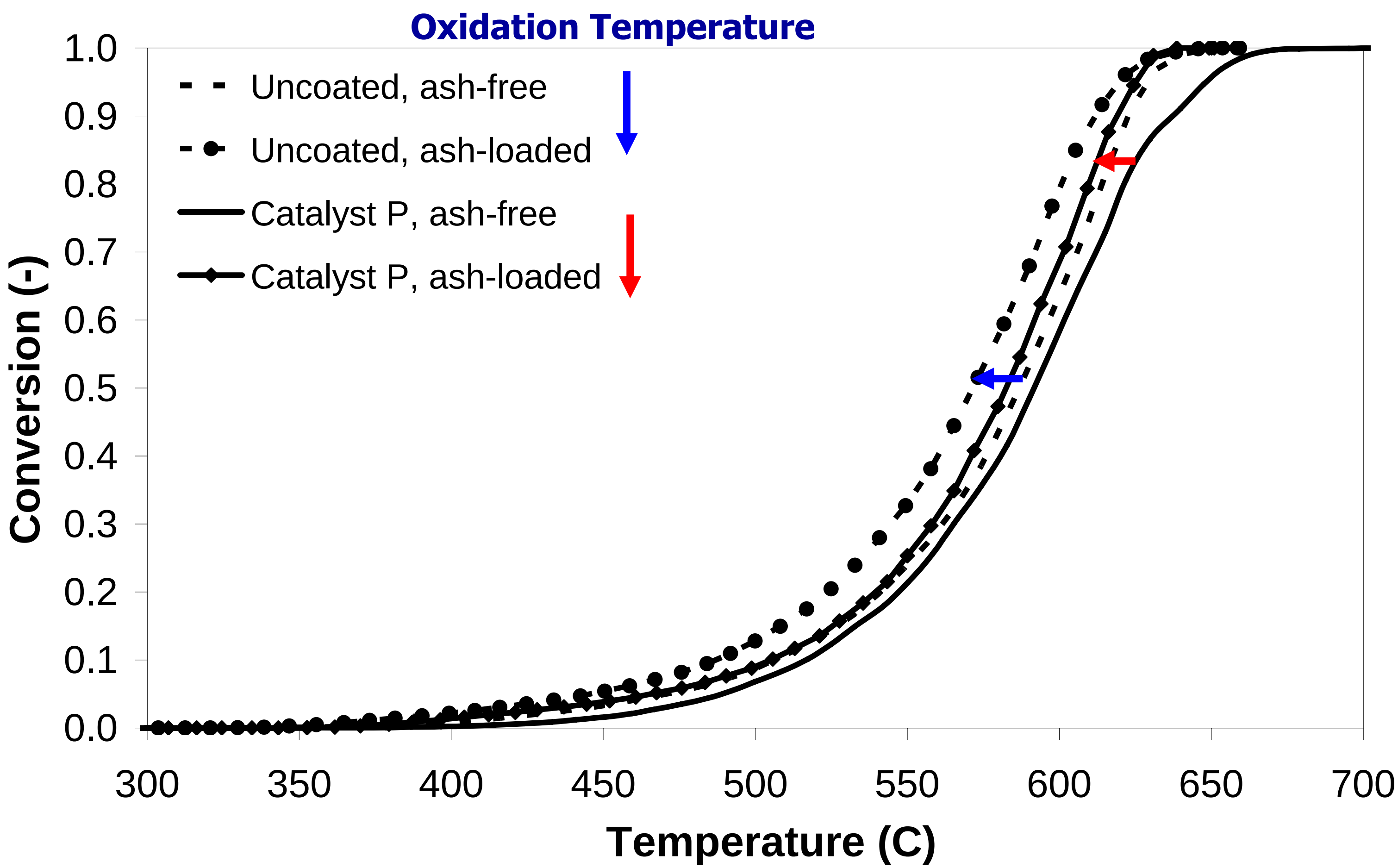
Soot Oxidation – Effect of the Catalyst



Catalyst M significantly enhances direct soot oxidation.

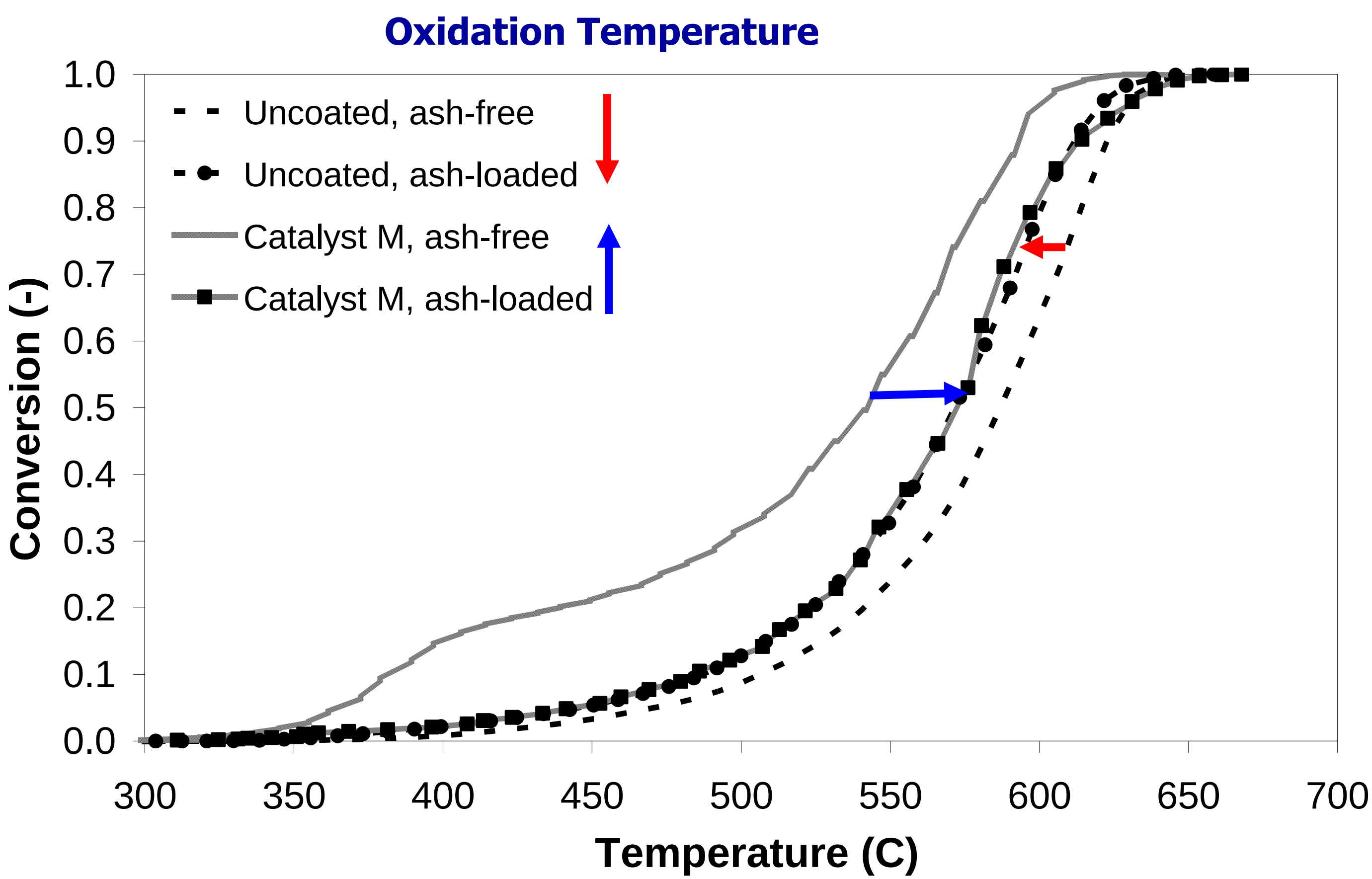
Catalyst P has no direct soot oxidation effect.

Filter Regeneration – Effect of Ash on Catalyst P



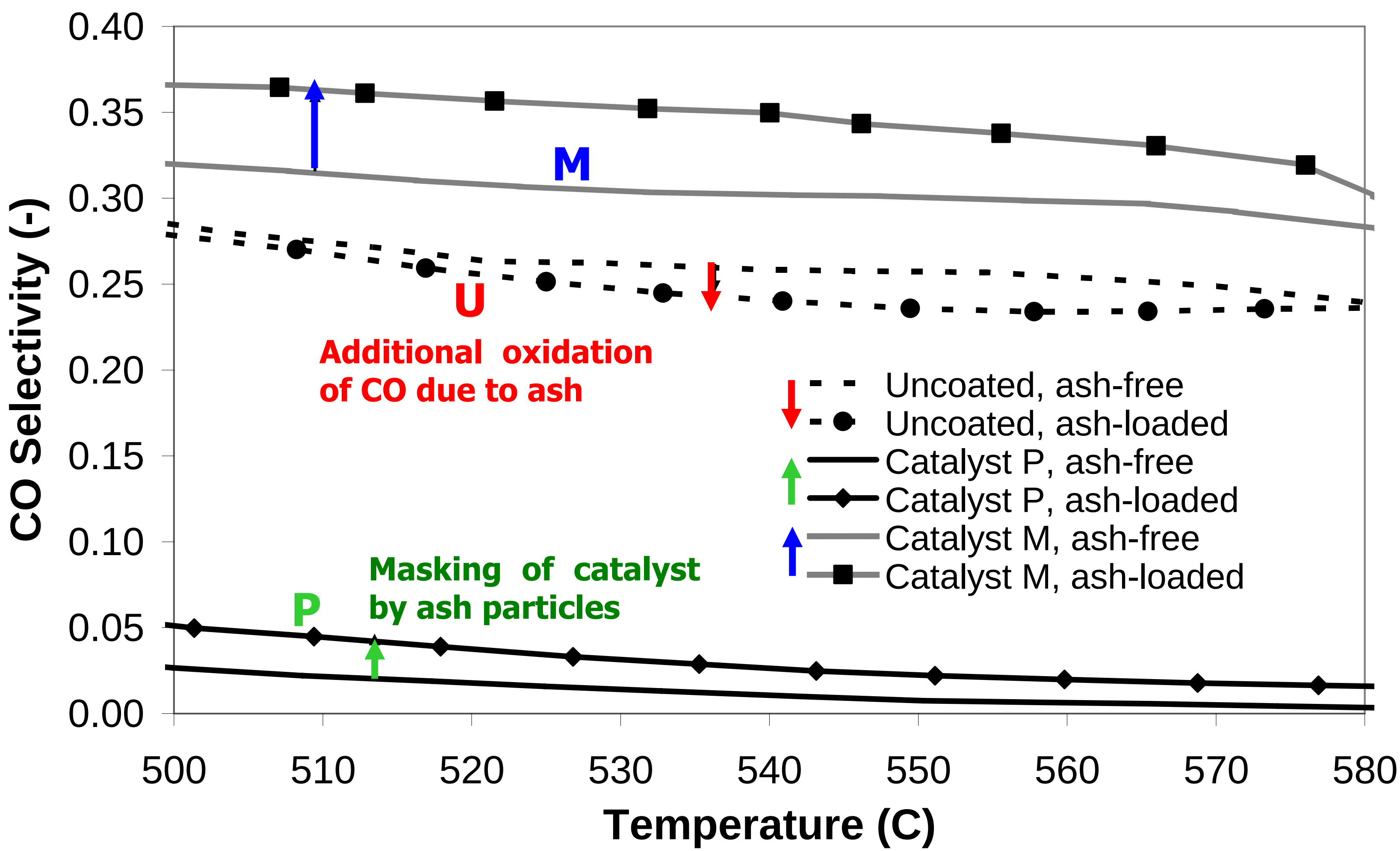
Small shift of the soot conversion curve to lower temperatures due to ash accumulation for both the uncoated filter and Catalyst P.

Filter Regeneration – Effect of Ash on Catalyst M



Significant loss of the catalytic activity of Catalyst M due to ash. The ash-loaded catalyzed filter oxidizes soot like the uncoated ash-loaded filter.

Filter Regeneration – Effect of Ash the CO Selectivity



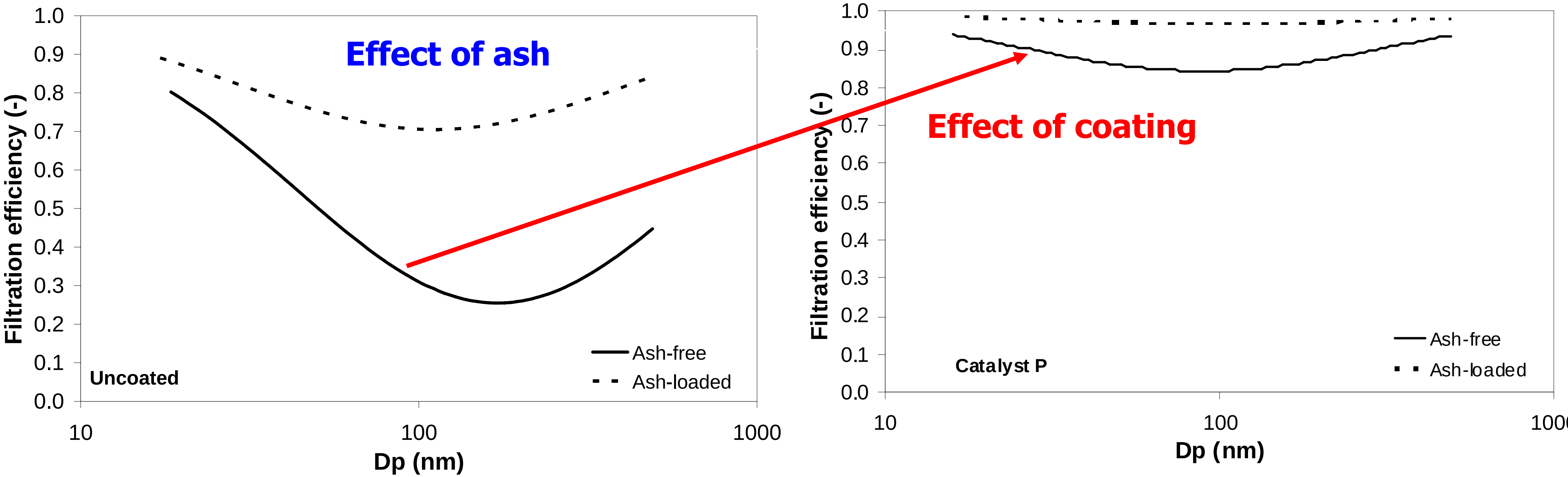
Effect of Ash on Filtration Efficiency

Uncoated filter:

Size-specific filtration efficiency – Effect of ash

Catalyst P filter:

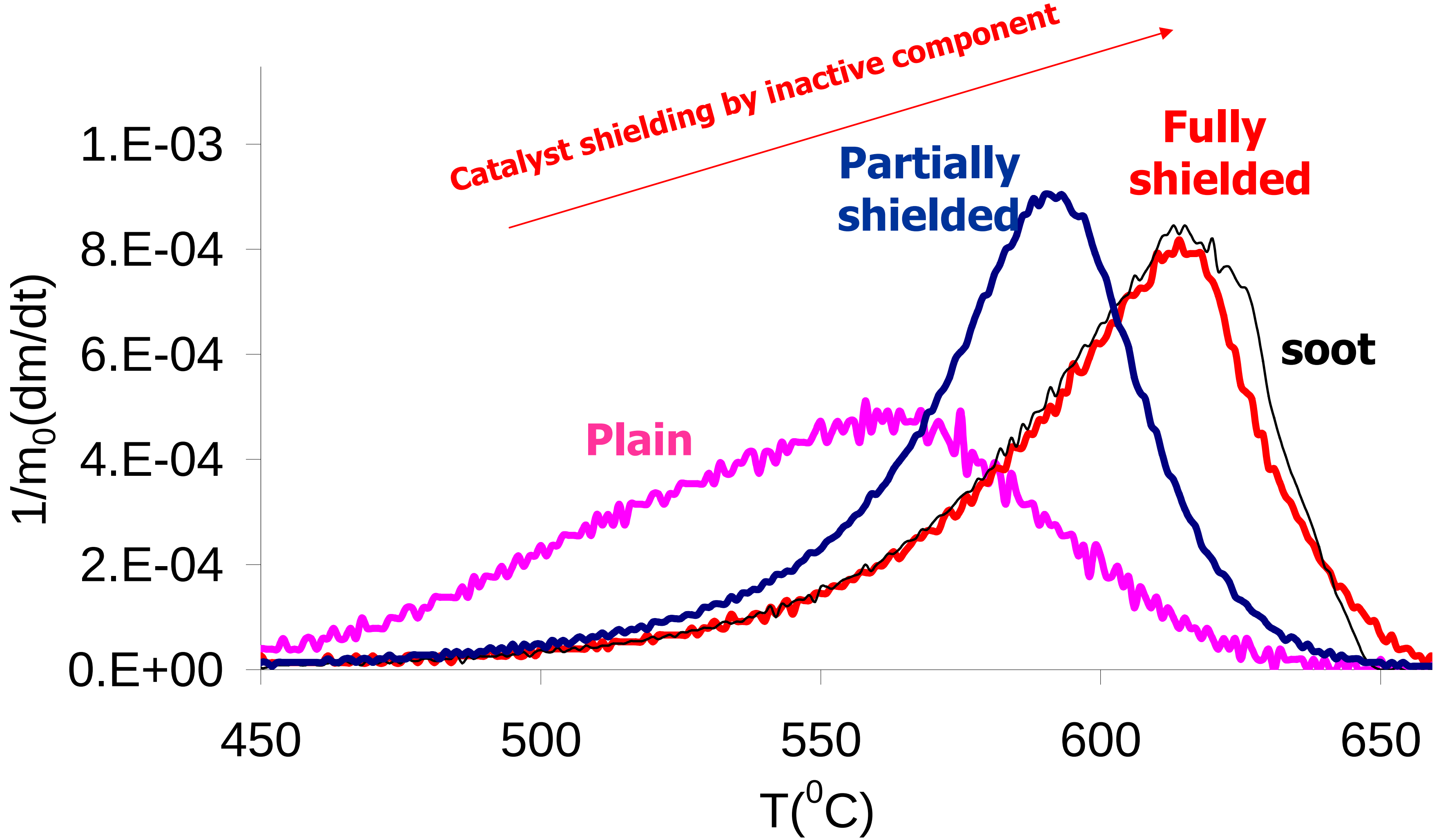
Size-specific filtration efficiency – Effect of ash



Significant increase in the filtration efficiency due to ash particle accumulation for both the uncoated and the catalyzed filter.

Similar results for catalyst M.

Current work: Effect of catalyst particle shielding by inactive components



Conclusions: Next Frontiers

- Contact micromechanics of soot aggregate-catalyst, soot mobility/restructuring
- Engineer catalyst particle substructure and reactivity with ash component tolerance
- Ash: Can we get rid of it?

Acknowledgements

- My colleagues from the APT Lab Evdoxia Kladopoulou, Alexandra Zygogianni, Georgia Kastrinaki, Souzana Lorentzou, Dimitrios Zarvalis, Akrivi Asimakopoulou Eleni Papaioannou, Nickolas Vlachos, Margaritis Kostoglou and Georgios Patrianakos
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