# Effect of Catalyst Coating on Pressure Drop and Efficiency of Ceramic Wall Substrate

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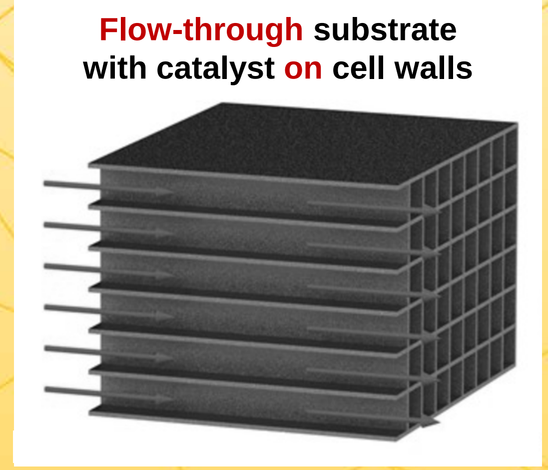
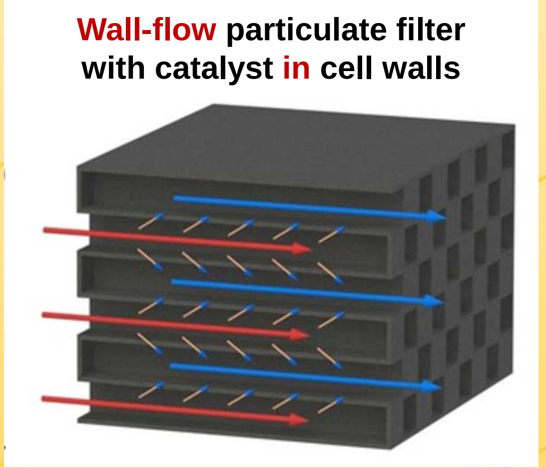
# Introduction – Catalyzed Particulate Filters

- Conventional vehicle emission control catalytic converters utilized monolithic flow-through supports coated with high surface area inorganic oxides (washcoat) and, in most cases, precious metals.



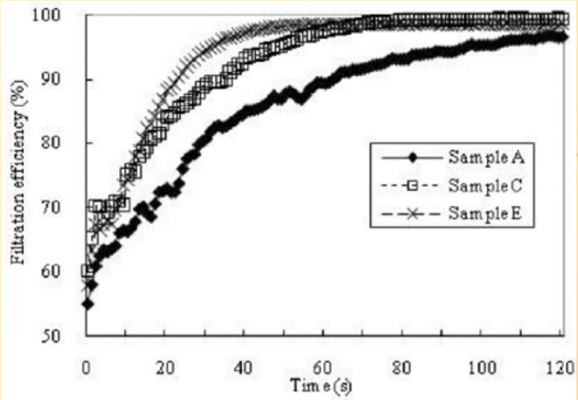




- When particulate emission control is required, a catalyst is applied onto the pores of a monolithic wall-flow substrate. The catalyzed filter can:
  - reduced space in the exhaust system;
  - lower the soot combustion temperature, allowing easier self-regeneration;
  - if coated with an oxidation catalyst, supply $NO_2$ for improving activity of a SCR catalyst;
  - If coated with an SCR catalyst, provide for faster light-off;
  - control particulate number emissions in DPF and GPF applications.

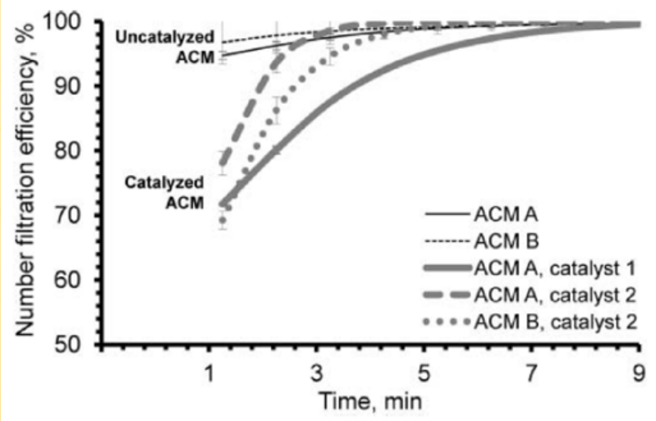
# Introduction – Effect of Catalyst on Filtration Efficiency

- Will addition of washcoat material into the ceramic microstructure improve soot removal efficiency?
  - Maybe Yes (Tsuneyoshi et al., 2011)

| Sample | A | B | C | D | E |
|---|---|---|---|---|---|
| Substrate | SiC | | | | |
| Size (mm) | Φ144 × L153 | | | | |
| Cell Density (cpsi) | 300 | | | | |
| Wall Thickness (mm) | 0.25 | | | | |
| Porosity (%) | 46.9 | 45.2 | 43.8 | 43.0 | 42.0 |
| Median Pore Diameter (μm) | 16.5 | 14.5 | 13.5 | 13.2 | 13.0 |
| Washcoat | none | yes | | | |
| Coat amount / Coat amount $_B$ | 0 | 1 | 1.4 | 1.7 | 2.1 |
| Pt Loading | none | | | | |




  - Maybe No (Swanson et al., 2013)




**<u>This is not a single case.</u>**

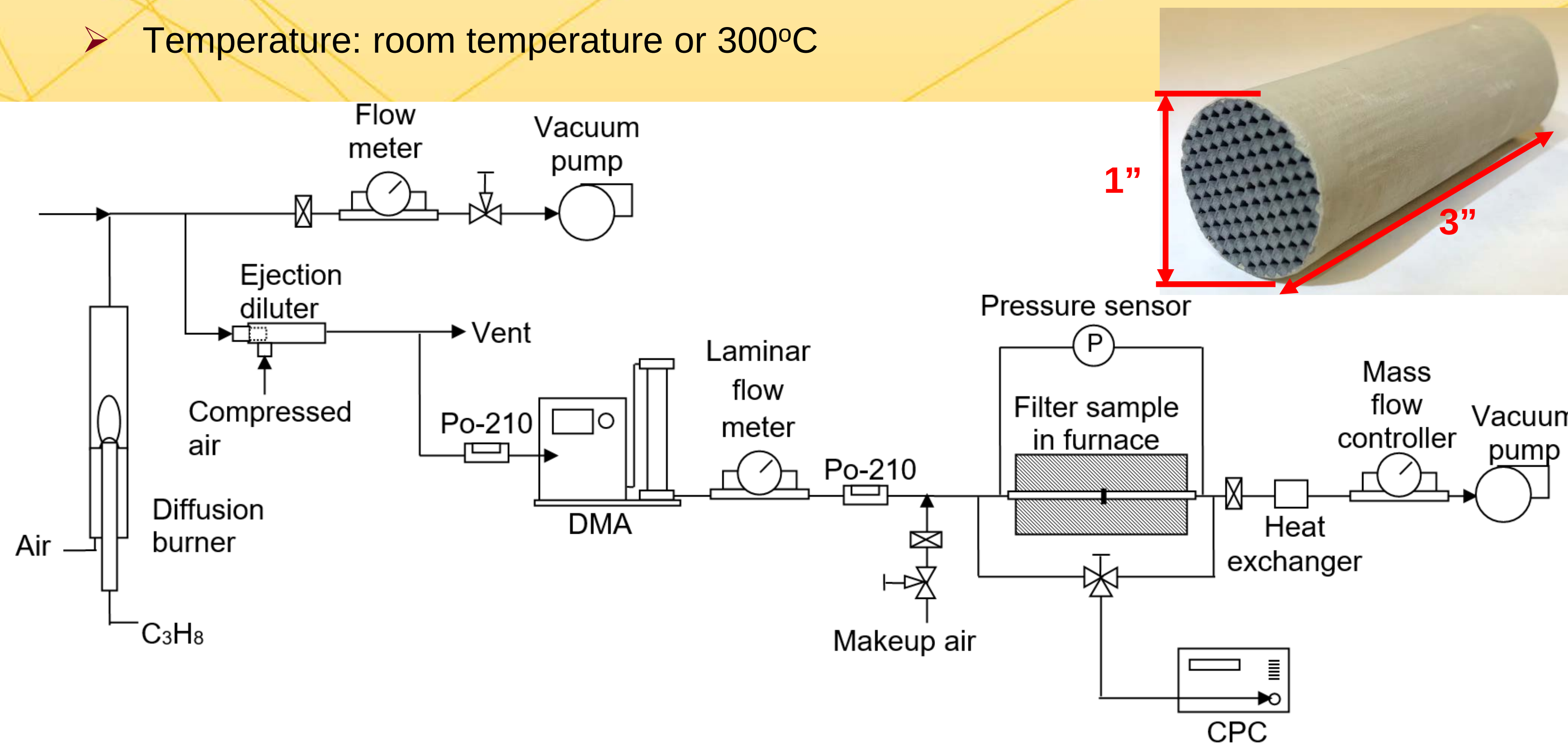
# Objectives

- **Evaluate and compare pressure drop and initial soot removal efficiency of ceramic wall flow substrates with and without catalyst coating.**

- **Identify the mechanistic causes, for change in pressure drop and initial filtration efficiency, as a result of application of catalytic washcoat into the filter structure.**

# Test Method

- Pressure drop and filtration efficiency of four blank (uncatalyzed) SiC wall-flow core samples (1” O.D. and 3” long) were measured using monodispersed soot particles from a home-made propane diffusion flame burner, at
  - Volumetric flow rate: 12.9 or 32.3 L/min (20,000 or 50,000 $h^{-1}$ space velocity)
  - Temperature: room temperature or 300$^{o}$C



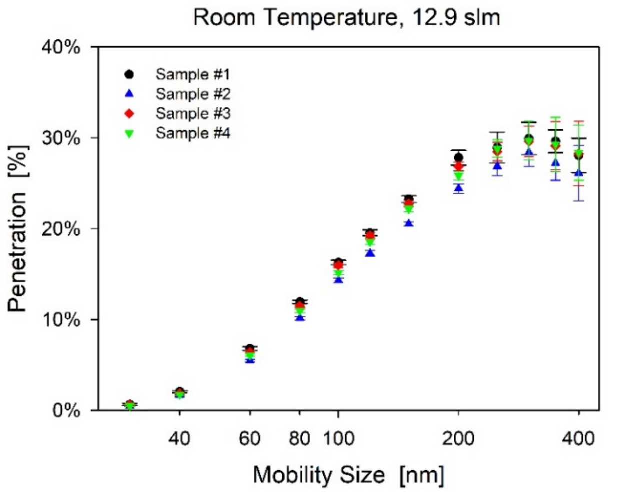
# Flow Resistance – Blank Filter

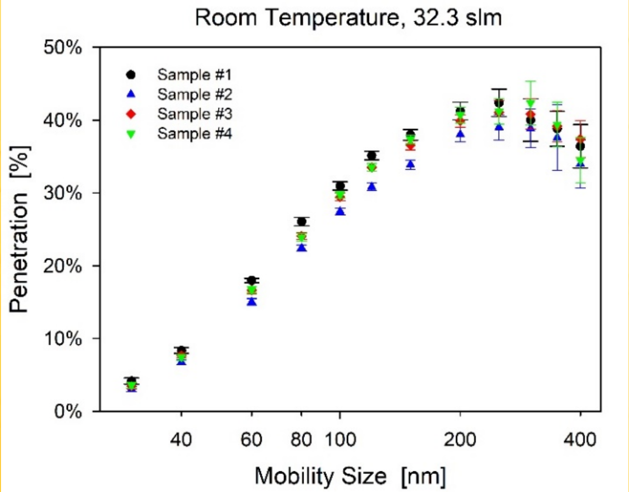
- At all test conditions, the pressure drop of the four filter core samples are very similar, indicating good consistency and uniformity.

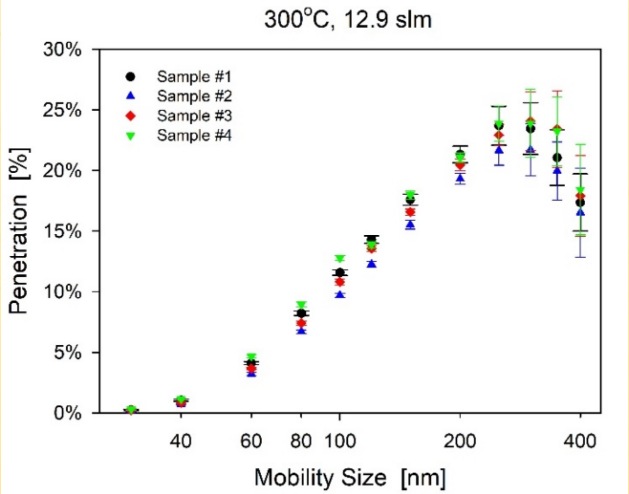
| Sample # | Pressure Drop [in $H_2O$] | | | |
|---|---|---|---|---|
| | Room temperature | | 300ºC | |
| | 12.9 slm | 32.3 slm | 12.9 slm | 32.3 slm |
| #1 | 0.26 | 0.71 | 0.55 | 1.14 |
| #2 | 0.25 | 0.69 | 0.54 | 1.12 |
| #3 | 0.25 | 0.69 | 0.53 | 1.13 |
| #4 | 0.25 | 0.68 | 0.54 | 1.13 |
| Mean | 0.25 | 0.69 | 0.54 | 1.13 |
| Std. | 0.00 | 0.01 | 0.01 | 0.01 |

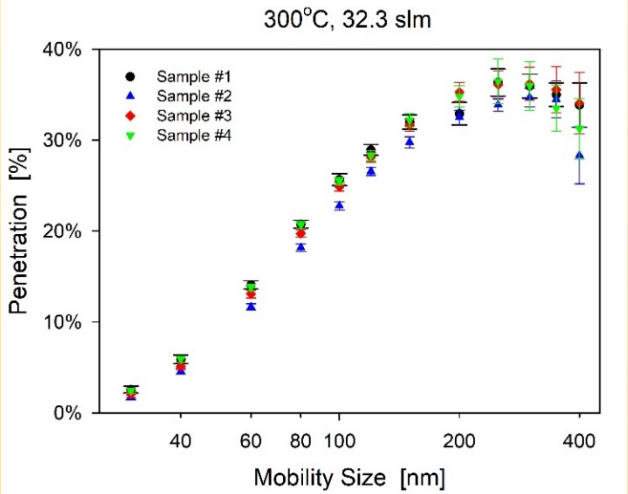
# Soot Removal – Initial Filtration Efficiency

- Similar soot fractional filtration efficiency was observed for the four samples. The largest variability is ±4% at high temperature and large particulate size.
- Good consistency in pressure drop and filtration efficiency suggests that this method can be used to evaluate the effect of washcoating, without concern from sample-to-sample variability.



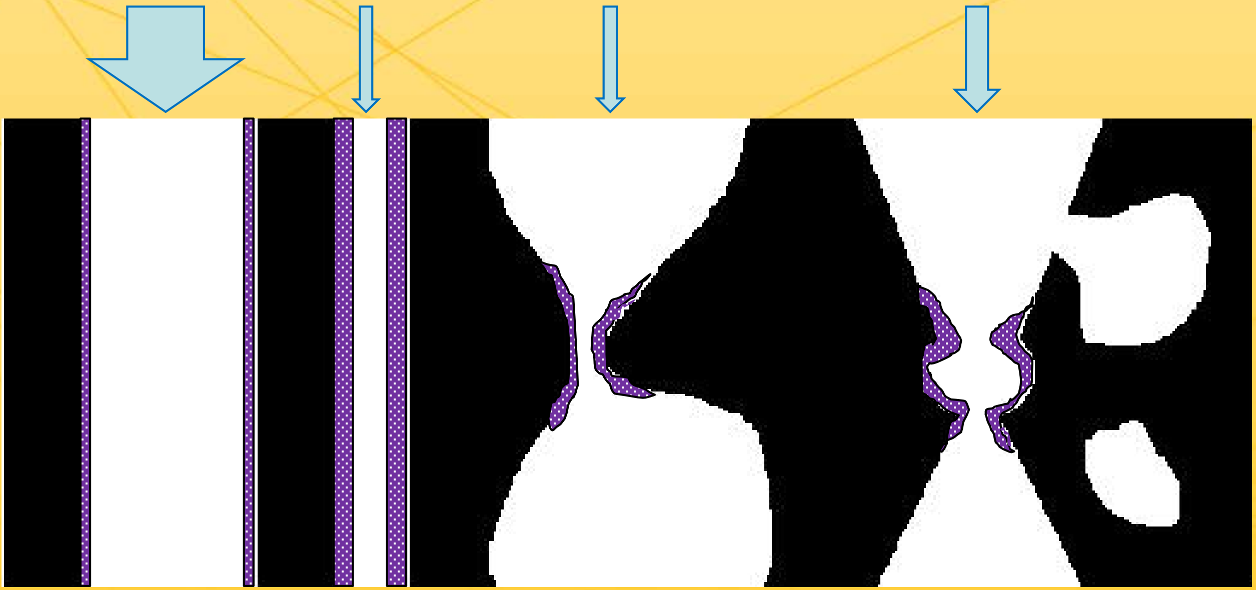



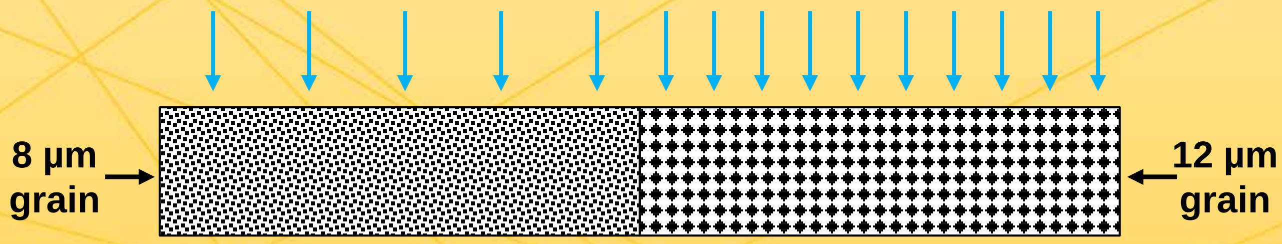
# Hypothesis for Reduced Filtration Efficiency – Pore Size Shift

- One possible explanation for reduced filtration efficiency after coating is media pore size distribution shift.
  - The capillary-driven washcoating process may preferentially block smaller pores, while leaving larger pores open.
  - Shift in pore size distribution induces more air flow towards larger pores, where more penetration occurs.



# Hypothesis for Reduced Efficiency –
## A Hypothetical Demonstration (I)

- If a ceramic substrate contains two grain sizes, 8 and 12 µm, which are completely separate and have equal area;
- after washcoating, X% of coating material is applied to the finer grains (8 µm), the rest is applied to the coarser grains (12 µm);
- coating material uniformly covers each grain surface, increasing effective grain size and solidity.

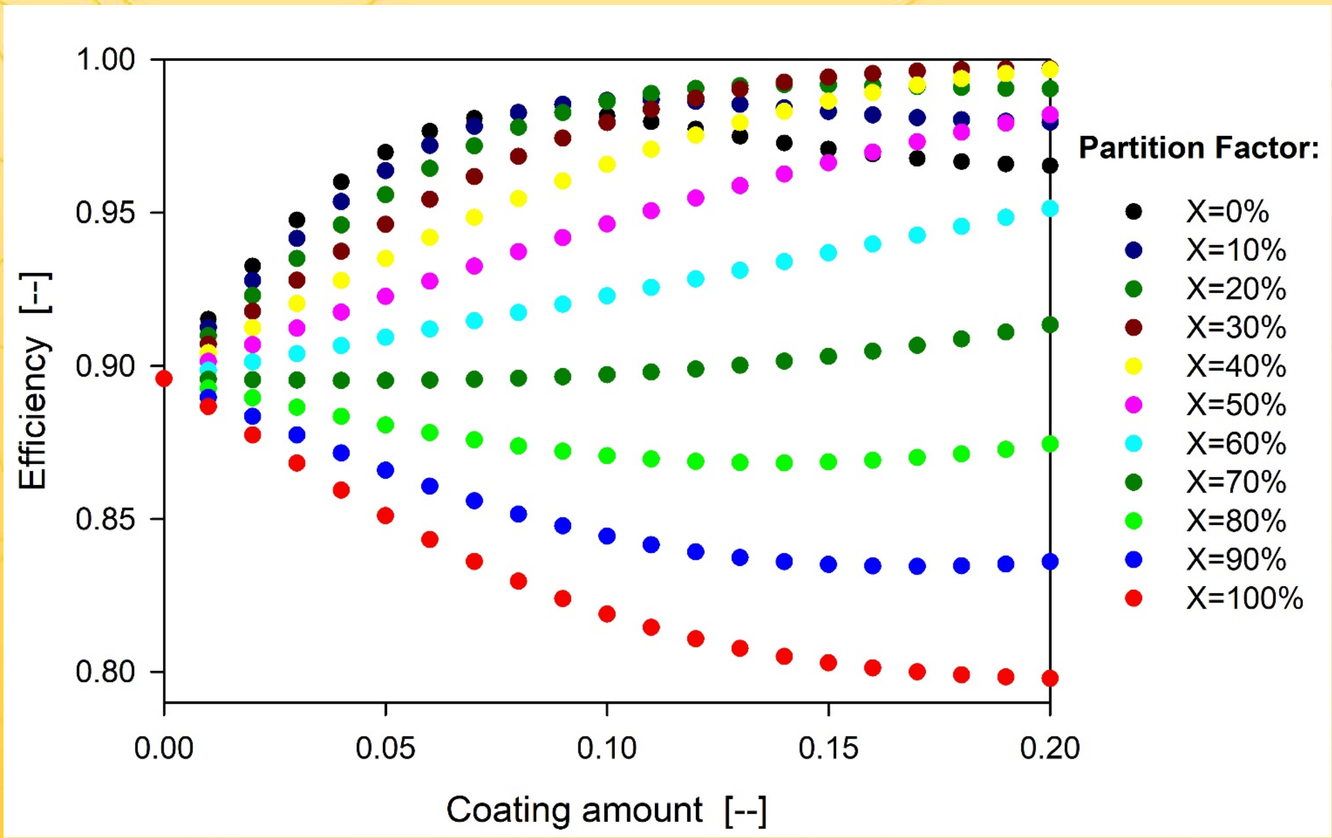



(See appendix for details)

| Coating amount [--] | Partition X [%] | Eff [%] | ΔP [Pa] | finer grain (8 µm) | | | | coarser grain (12 µm) | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | $\alpha_1$ [--] | $d_{c1}$ [µm] | $U_1$ [cm/s] | $Eff_1$ [%] | $\alpha_2$ [--] | $d_{c2}$ [µm] | $U_2$ [cm/s] | $Eff_2$ [%] |
| Blank | **/** | 89.6 | 566 | 0.5 | 8.0 | 1.8 | 99.6 | 0.5 | 12.0 | 4.0 | 85.1 |
| 0.1 | **0** | 98.2 | 1382 | 0.5 | 8.0 | 4.3 | 97.7 | 0.7 | 13.4 | 1.4 | 99.6 |
| 0.1 | **50** | 94.6 | 1351 | 0.6 | 8.5 | 1.8 | 99.9 | 0.6 | 12.8 | 4.0 | 92.3 |
| 0.1 | **100** | **81.9** | **768** | 0.7 | 9.0 | 0.4 | 100.0 | 0.5 | 12.0 | 5.4 | 80.7 |

# Hypothesis for Reduced Efficiency –
## A Hypothetical Demonstration (II)

- If the wachcoat partition among different sized pores varies, the filtration efficiency of the coated wall-flow substrate could reduced even though the overall porosity decreases.

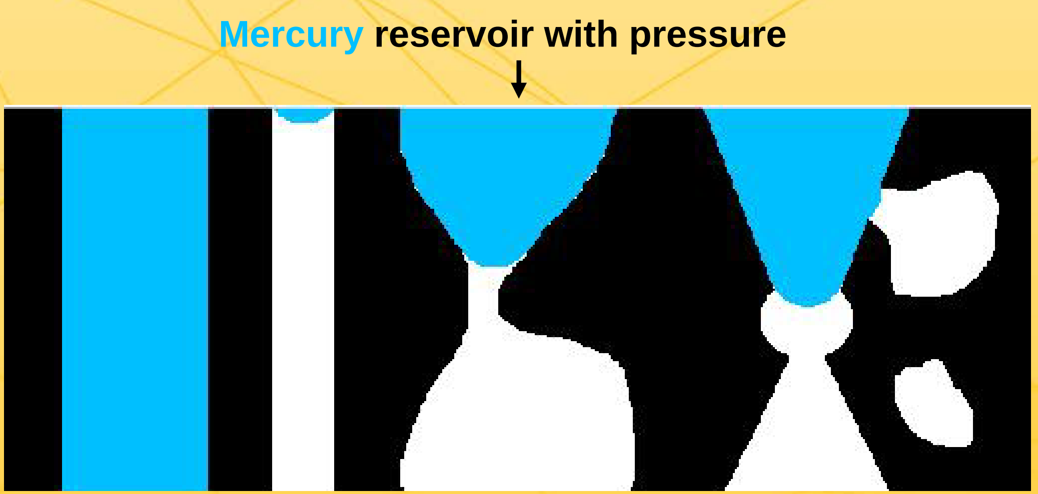


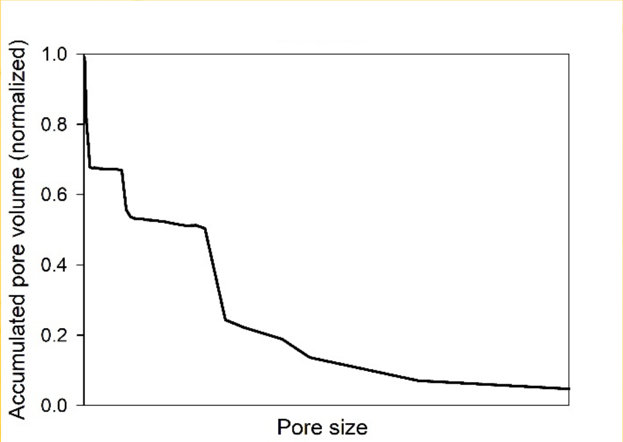
# Pore Size Distribution Measurement
## – Mercury Intrusion Porosimetry

- Mercury intrusion porosimetry is one of the most widely used techniques to measure the pore size distribution of porous media:
  - Pressure required to introduce a non-wetting liquid into a pore is associated with an equivalent pore diameter:

$$P = 4\gamma \cos\theta / D$$

$P$: pressure applied
$\gamma$: liquid surface tension
$\theta$: liquid-solid contact angle
$D$: equivalent diameter



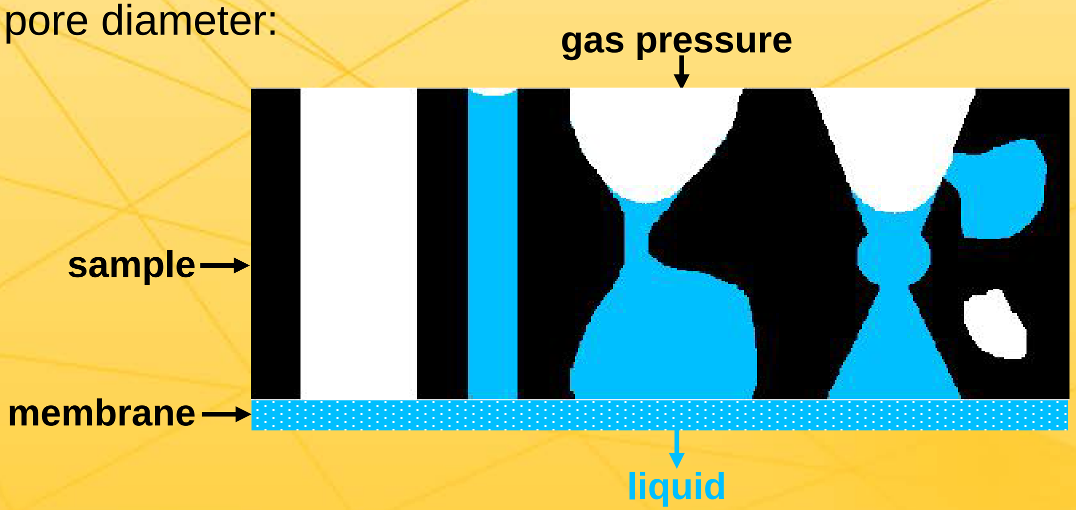




- Both through pores and blind pores are measured;
- Pore volumes are measured partially. No flow characteristics of pores are measurable.

# Pore Size Distribution Measurement

## – Liquid Extrusion Porosimetry

- In liquid extrusion porosimetry, a pressurized non-reacting gas is used to remove liquid from pores of the sample:
  - A membrane whose largest pore is smaller than the smallest pore of interest in the sample is placed underneath the sample
  - A liquid that wets the sample and the membrane is used to fill all the pores of the sample and the membrane;
  - Pressure required to displace the wetting liquid in a pore is associated with an equivalent pore diameter:



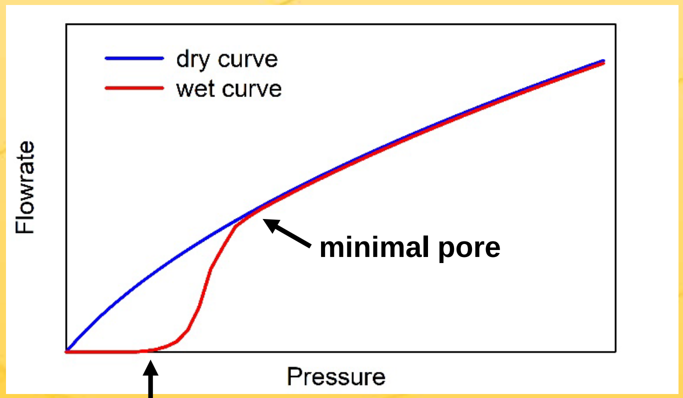

- Only through pores are measured. (results not interferred by blind pores)
- Pore volumes are measured partially. No flow characteristics of pores are measurable.

# Pore Size Distribution Measurement
## – Extrusion Flow Porometry

- Extrusion flow porometry (capillary flow porometry) is a characterization technique based on the displacement of a wetting liquid from the sample pores by applying an inert gas at increasing pressure.
  - ➢ Presence of pores is detected by sensing the increase in gas flowrate at any given differential pressure, which corresponds to pore size.

gas flow

gas pressure




bubble point

❖ Only through pores are measured. (results not interferred by blind pores)

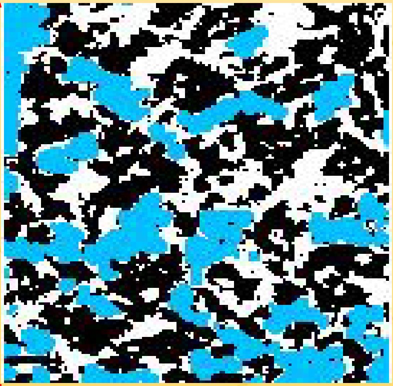
❖ Pore volumes are not measured. Flowrate through each pore size is measurable, which may be particularly relevant to the filtration process.

# Pore Size Distribution Measurement

## – 3D structure reconstruction

- 3-D micro-structures from images captured through various techniques (e.g., X-ray µCT or FIB-SEM) can enable calculation of pore size distributions from digital scan images, if pore size characterization tool is not available.

(An example layer of a SiC wall from X-ray µCT)



(An example layer of a SiC wall with simulated mercury intrusion)

- Potential analysis from 3-D structure:
  - Intrusion and extrusion porosimetry (geometric pore size with connectivity consideration)
  - Extrusion capillary porometry and permeability (lattice Boltzmann or Navier-Stokes)
  - Filtration efficiency based on particle tracking (Navier-Stokes with Lagrangian specification for particle movement)

# Summary

- Pressure drop and soot removal efficiency of four blank SiC wall-flow core samples (1” O.D. and 3” long) were measured at different flowrates and temperatures.
- Good consistency was found among the samples, which will enable comparison between blank and coated filter cores.
- Filtration performance of coated filter cores will be evaluated, and compared with the reference blank samples, in order to quantify the effect of coating methods and coating quantity.
- A possible explanation for reduced filtration efficiency after coating is proposed, based upon a shift in pore size distribution which changes the flow distribution through pores. The hypothesis is further illustrated with a hypothetical model.
- A review of pore size distribution measurement techniques suggests that extrusion flow porometry can serve as a basic characterization tool, and 3-D structure characterization can provide further details of porous wall micro-structure.

# Appendix

- Equations used for illustration in slide 9 & 10:
  - Pressure drop of a wall substrate with uniform spherical grain size:

$$\Delta P = \frac{18\mu Uth\alpha\left(3+2\alpha^{5/3}\right)}{\left(3-4.5\alpha^{1/3}+4.5\alpha^{5/3}-3\alpha^{2}\right)d_s^2},$$

(Rudnick and First, 1978)

  - Collection efficiency of a wall substrate with uniform spherical grain size:

$$P = \exp\left(\frac{-3\alpha Et}{2d_s\left(1-\alpha\right)}\right), \qquad E = 1-(1-E_D)(1-E_R).$$

$$E_D = 3.5\left(\frac{1-\alpha}{Ku}\right)^{1/3} Pe^{-2/3}; \qquad E_R = 1.5\left(\frac{1-\alpha}{Ku}\right)\frac{R^2}{\left(1+R\right)^{(1+2\alpha)/(3-3\alpha)}}.$$

(Kuwabara, 1959)

(Tardos et al., 1976)

  - Change in grain size after coating:

$$\frac{\alpha_{blank}}{\alpha_{coated}} = \frac{d^3_{s\_blank}}{d^3_{s\_coated}}.$$

  - Ensuring same differential pressure across 8μm and 12μm wall.

$$\Delta P_{8\mu m} = \Delta P_{12\mu m}.$$

| | | | |
|---|---|---|---|
| $\Delta P$: | pressure drop | $P$: | penetration |
| $\mu$: | air viscosity | $E$: | single sphere efficiency |
| $U$: | face velocity | $E_D$: | single sphere efficiency by diffusion |
| $th$: | wall thickness | $E_R$: | single sphere efficiency by interception |
| $\alpha$: | packing density | $Ku$: | Kuwabara flow constant for sphere bed |
| $d_s$: | grain size | $Pe$: | Péclet number |
| | | $R$: | interception parameter |