Measurement and Modelling of PM Loading in Bare and Catalytic Flow-Through Monoliths

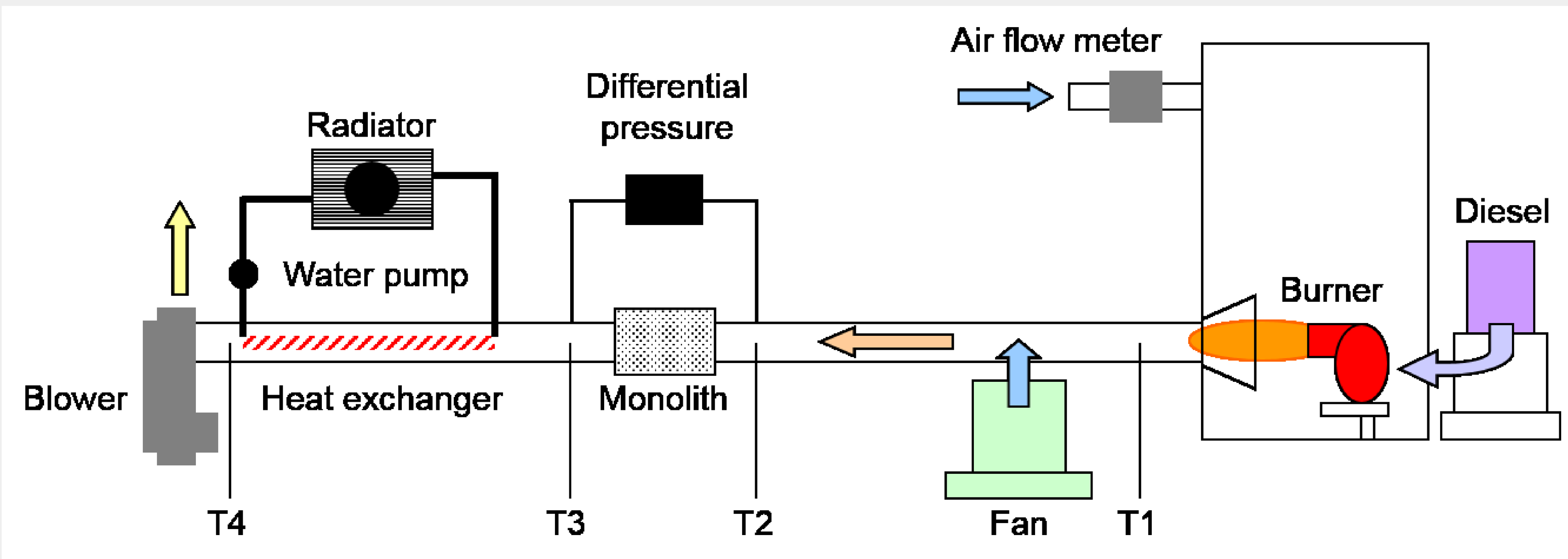
Onoufrios Haralampous*, Simon Payne

UNIVERSITY OF CAMBRIDGE
Department of Engineering

Introduction

Particle collection mechanisms at the leading edge and in open channels of honeycomb monoliths are usually neglected by engineers of these devices. Under specific conditions however, these phenomena can make an appreciable contribution to overall particle collection and deposits on channel walls may affect catalyst performance. Deposit loading tests conducted with a Diesel Particle Generator are used in this study to highlight the differences in capture efficiency and deposit evolution between bare and catalytic flow-through monoliths. A recently developed diesel particle collection model is extended to take into account the diffusion of active gaseous species in the deposit layer and their reaction with soot.

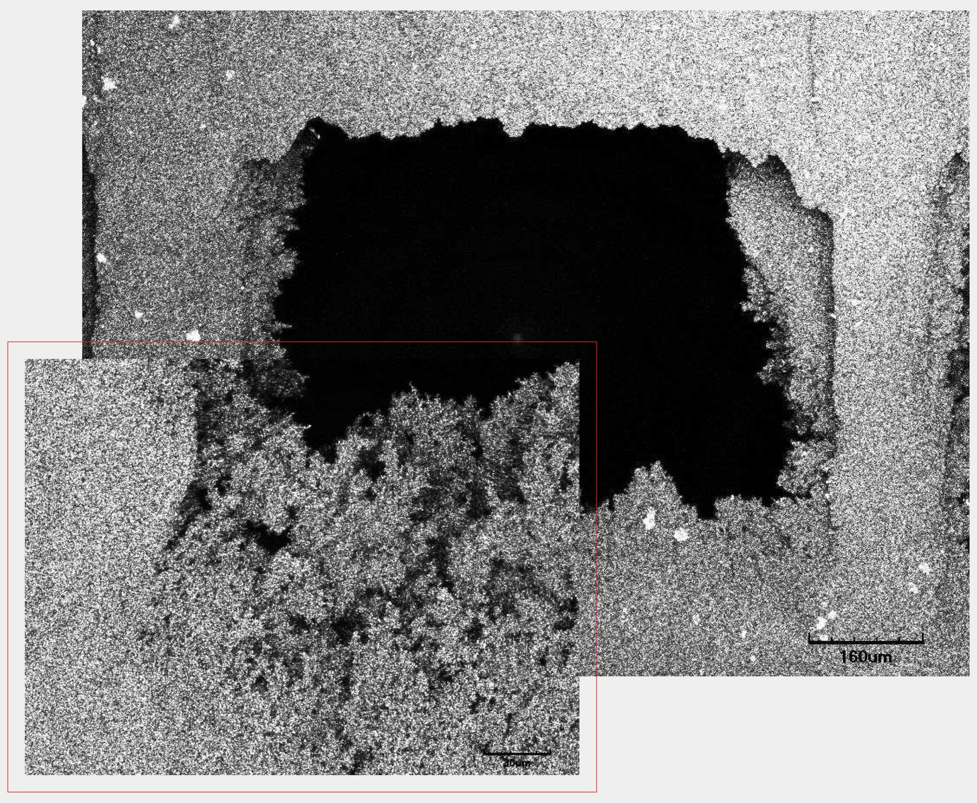
Experimental testing



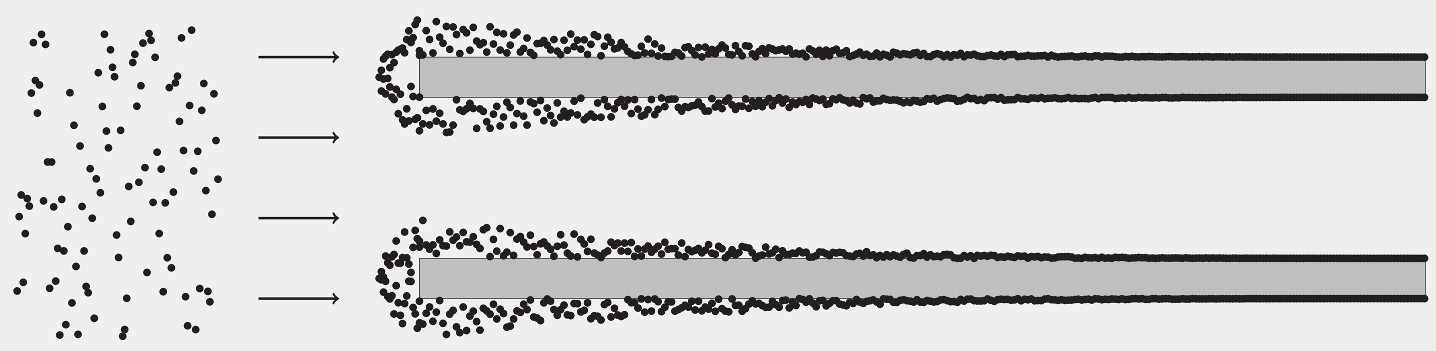
- In previous work at the University of Cambridge, long-term loadings of flow-through monoliths were carried out using prototype and commercial versions of a Diesel Particle Generator (DPG) [1].

	Varied parameters
Monolith design	cell density, wall thickness, length, coating
Operating condition	particle size, loading rate and duration, flow rate

- Measurements included particle emissions, monolith pressure drop, deposit mass (edge, channel distribution).
- Confocal laser microscopy images were used to study the deposit micro-structure.



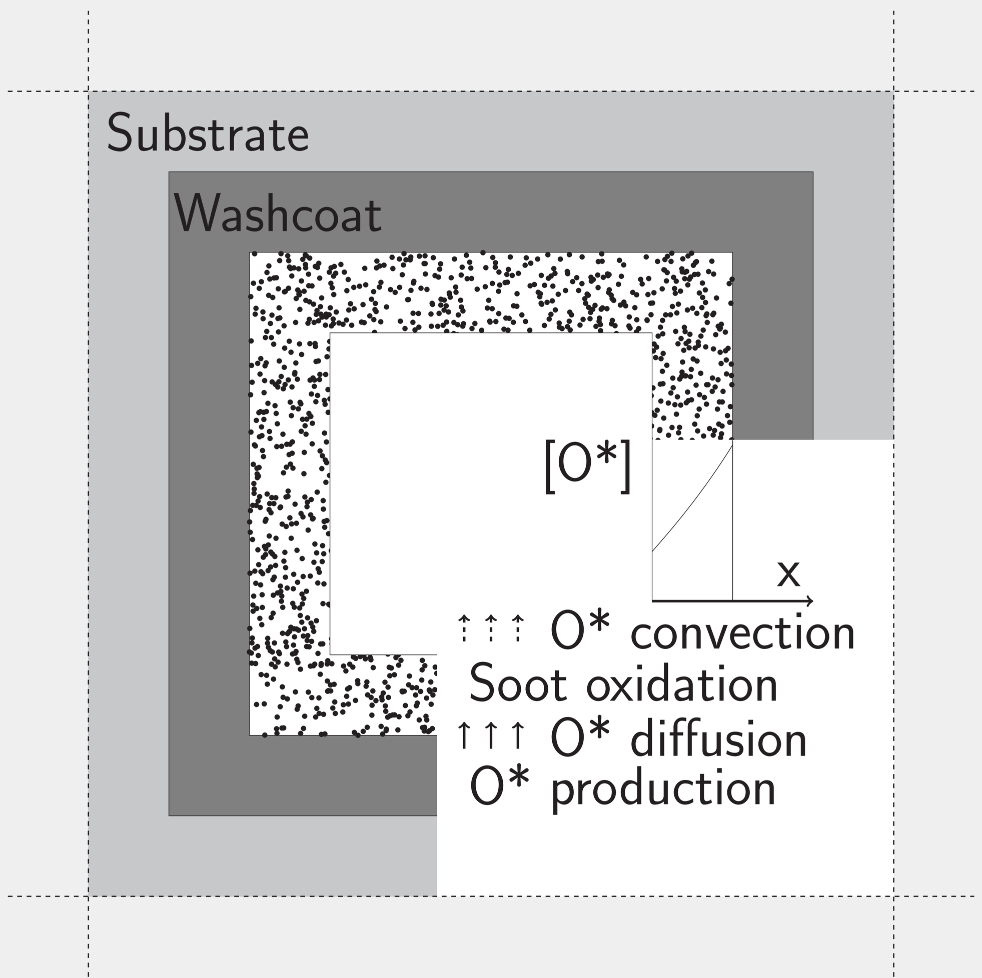
Mathematical modelling



Interception at leading edge

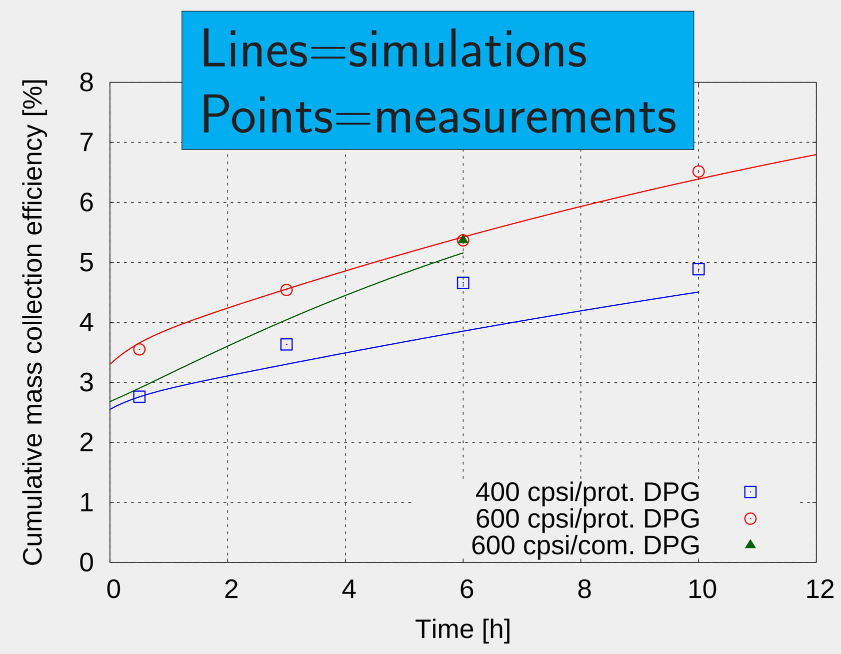
Coupled diffusion-interception inside channel

- PM collection model[2]
  - 1-d axial discretization & multiple particle classes
  - Local collection efficiency as a function of particle size & deposit buildup in dendrite form
- Loose-contact catalytic soot oxidation
  - Oxygen spillover as described in [3]
  - Calculation of activated oxygen (O*) concentration profile



Bare monolith results

- Measured parameters as a function of time
  - Cumulative collection efficiency
  - Pressure drop
  - Deposit distribution
- All trends are predicted with sufficient accuracy



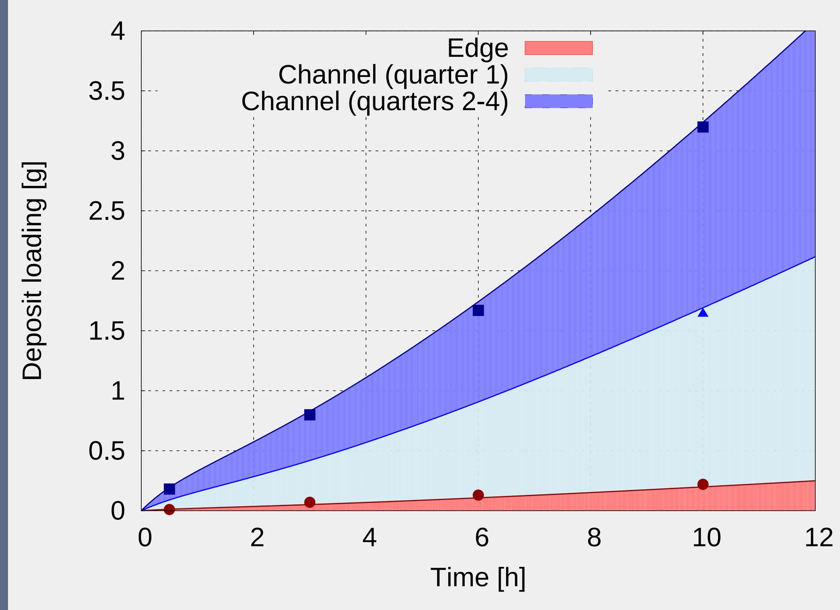
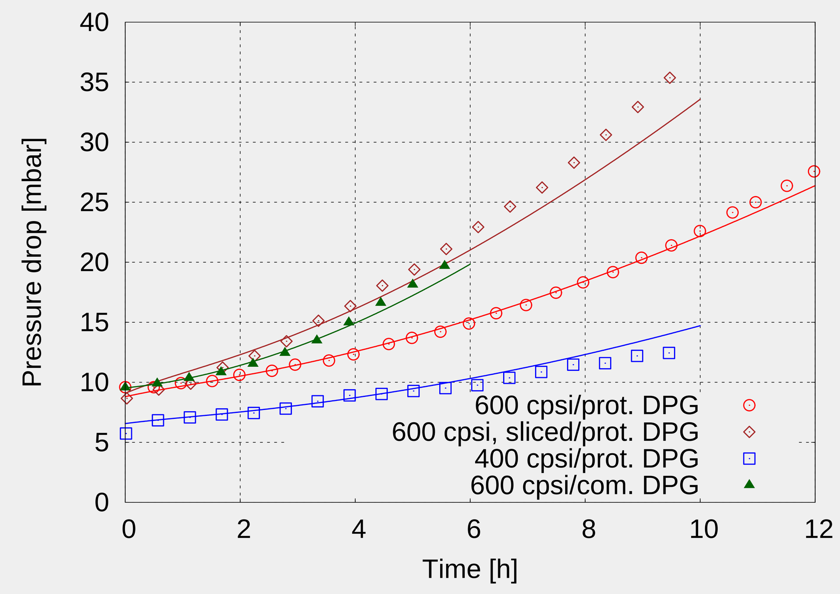


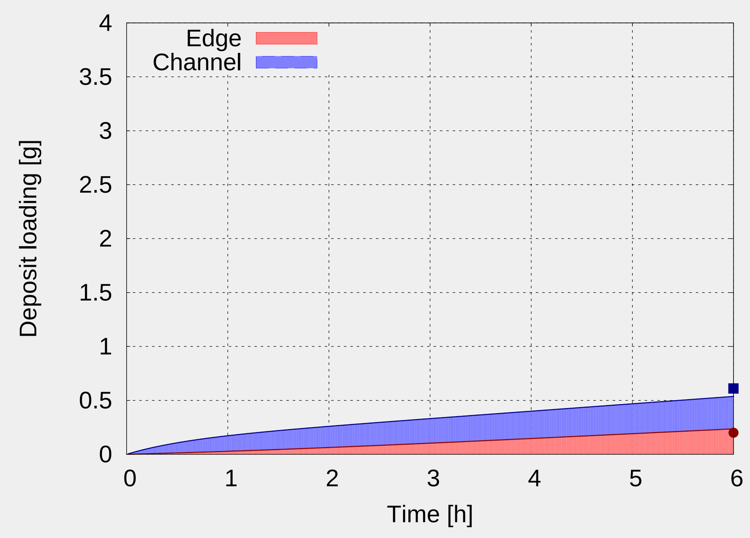
Figure: Deposit distribution compiled from a series of 5 tests for the 600 cpsi monolith loaded on the prototype DPG.

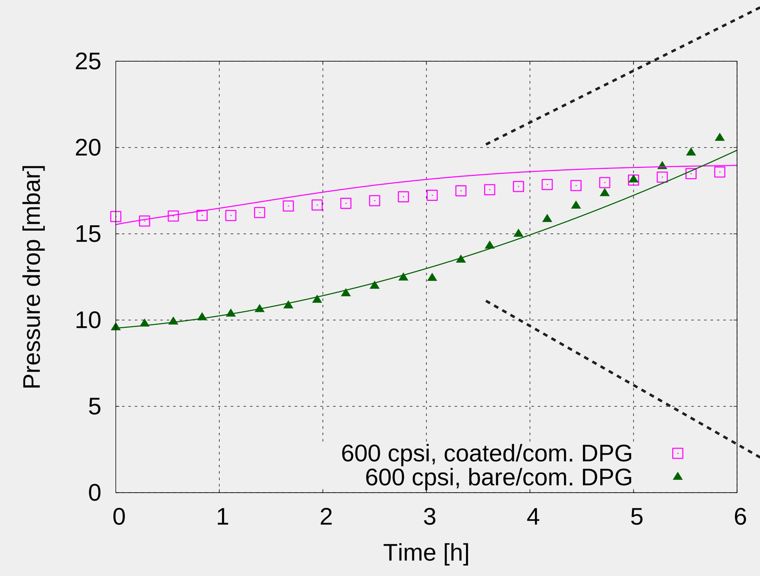


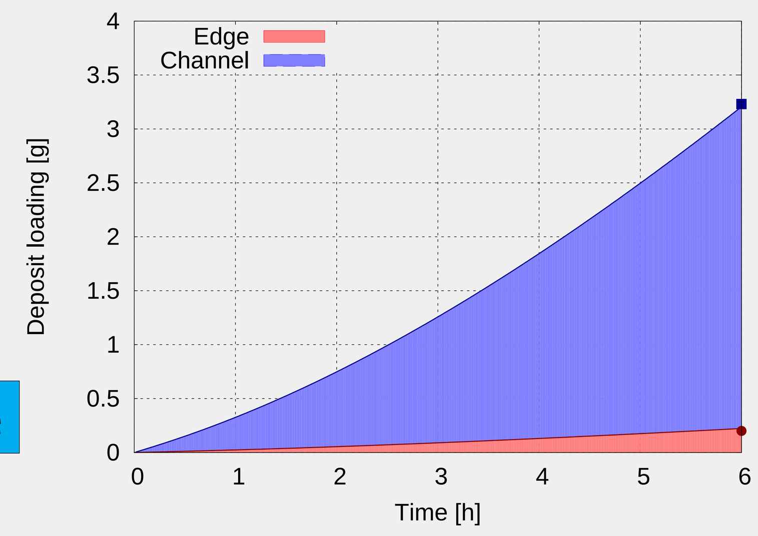
Effect of catalytic coating

- Very good prediction of both pressure drop and loading evolution
- Insignificant effect of coating on collection efficiency at clean state (=2.7%)
- Cumulative PM collection efficiency after 6h = 3.4% (vs 5.1% for bare)
- 58% of deposit collected in channel is oxidized according to the model
  - loose contact mechanism (insignificant oxidation with NO_2)
- No oxidation of deposit collected at the leading edge

Coated with $Pt-Pd/Al_2O_3$







Commercial DPG
20% O_2, 4ppm NO_x
$T = 240°C,\ \dot{m} = 200 kg/h$

Bare

Outlook and conclusions

- The effect of coating on PM collection in flow-through monoliths was studied experimentally and theoretically using a specially developed model
- 87% less mass is collected in the channel as a result of lower collection efficiency and catalytic oxidation of soot
- The buildup of deposit at the leading edge is not affected by coating
- Significant initial backpressure penalty but minimal increase during loading

References

Payne S. D., Experimental studies of diesel particle filtration, PhD Dissertation, *University of Cambridge*, UK, **2011**

Haralampous O., Payne S., Experimental Testing and Mathematical Modelling of Diesel Particle Collection in Flow-Through Monoliths, *Journal of Engine Research*, DOI: 10.1177/1468087416637735, **2016**

Zeng L., Weber A. P., Transportmechanismen für aktivierten Sauerstoff in der katalytischen Rußoxidation an Nanopartikel-Schichtsystemen, *Chemie Inginieur Technik*, **2012**, 84, No. 3, 295-300

* TEI of Thessaly, Mechanical Engineering Department, 41110, Larissa, Greece   onoufrios@teilar.gr

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Onoufrios Haralampous*[1] and Simon Payne[2]

[1]Technological Educational Institute of Thessaly, Mechanical Engineering Department
[2]University of Cambridge, Department of Engineering

1 Introduction

The honeycomb monolith is the most prevalent geometry in automotive exhaust aftertreatment, with applications including oxidation catalysts, partial-flow and wall-flow filters. Particle collection mechanisms at the leading edge and in open channels of honeycomb monoliths are usually neglected by engineers of these devices. Under specific conditions however, these phenomena can make an appreciable contribution to overall particle collection and deposits on channel walls may affect catalyst performance. Deposit loading tests conducted with a Diesel Particle Generator are used in this study to highlight the differences in capture efficiency and deposit evolution between bare and catalytic flow-through monoliths. A recently developed diesel particle collection model is extended to take into account the diffusion of active gaseous species in the deposit layer and their reaction with soot.

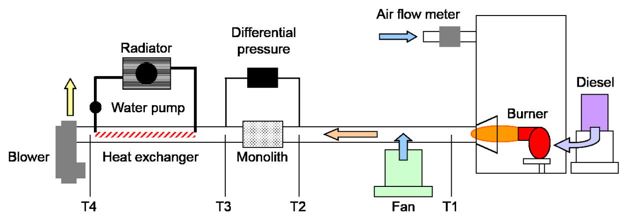


Figure 1: Schematic of prototype Diesel Particle Generator

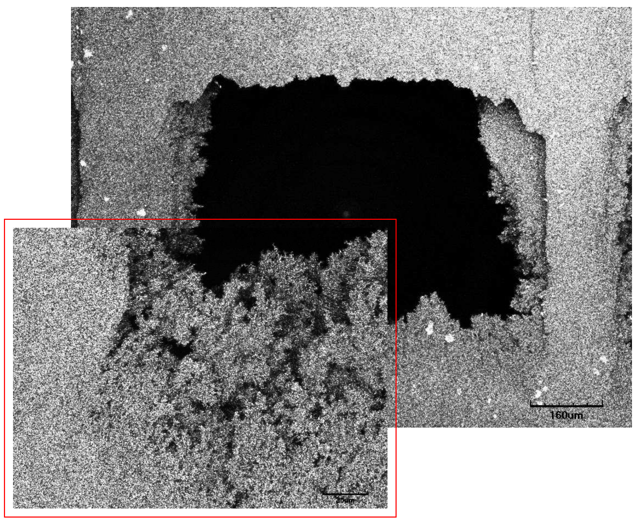


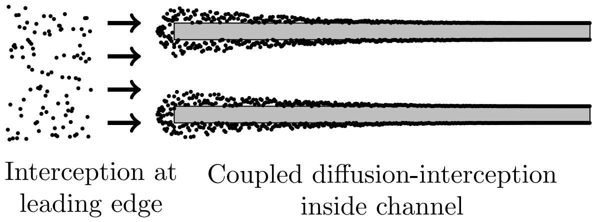
Figure 2: Confocal laser microscopy images of deposit microstructure

2 Experimental testing

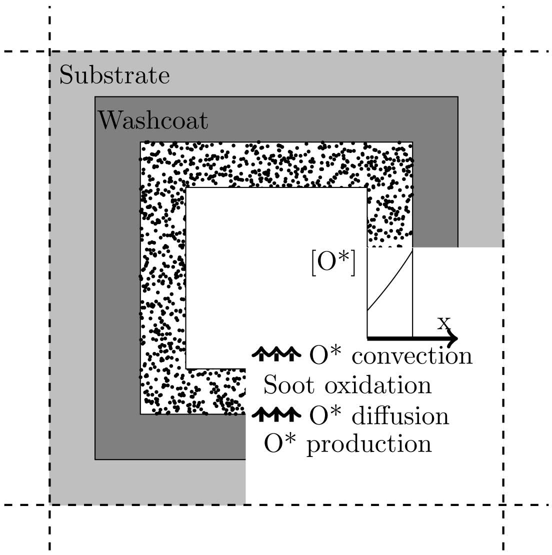
In previous work at the University of Cambridge, long-term loadings of flow-through monoliths were carried out using prototype and commercial versions of a Diesel Particle Generator (DPG) (1). Four monolith designs were used to study the effect of cell density, wall thickness, and coating. One of these monoliths, was a sliced and assembled in four pieces

*Corresponding author, onoufrios@teilar.gr in order to evaluate the effect of length and turbulence intensity. Furthermore two versions of the DPG were used covering different operating conditions in terms of particle size, flow rate, loading rate and duration. Measurements included particle emissions, monolith pressure drop, deposit mass (edge, channel distribution), while digital and confocal laser microscopy images (Fig. 2) were used to study the deposit micro-structure.

3 Mathematical model



(a) PM collection mechanisms



(b) Loose contact catalytic soot oxidation

Figure 3: Basic modelling assumptions

A recently published model(2) was used to simulate the tests. 1-d axial discretization and multiple particle classes were employed to predict local collection efficiency as a function of particle size and deposit buildup in dendrite form. The model was further extended with a loose-contact catalytic soot oxidation mechanism in accordance with the oxygen spillover described in (3). The mechanism comprised of the following steps: (1) activated oxygen (O^*) production on the catalytic surface (2) Brownian diffusion of O^* in the deposit layer and (3) oxidation of deposit with O^*.

4 Bare monolith results

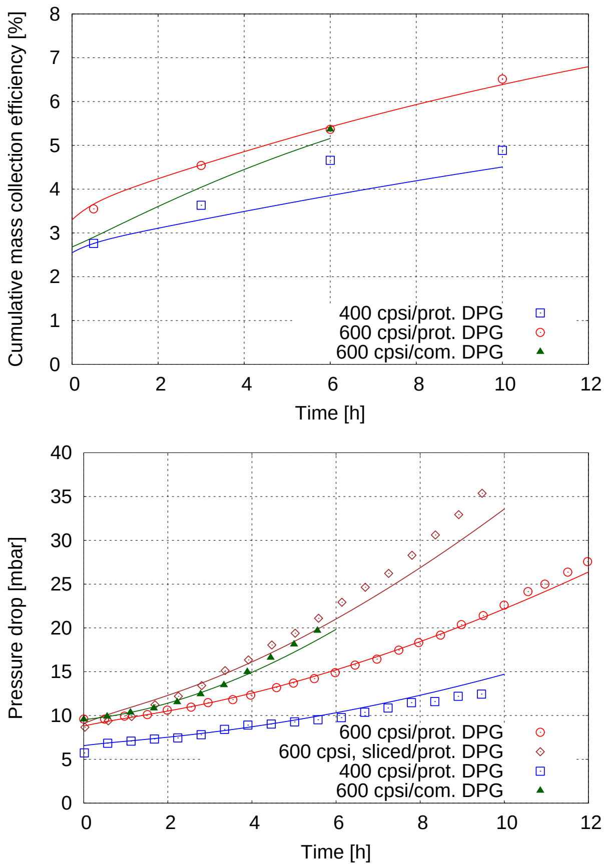


Figure 4: Performance of tested monoliths as a function of time (lines=simulations, points=measurements).

The model predicted with sufficient accuracy the evolution of cumulative collection efficiency, pressure drop and deposit distribution, as shown in Fig. 4.

5 Effect of catalytic coating

A 600 cpsi monolith, coated with $Pt-Pd/Al_2O_3$, was loaded on the commercial DPG ($T = 240°C$, $\dot{m} = 200kg/h$) to assert the effect of catalytic coating. As shown in Fig. 5, the extended model was successfully employed to predict the pressure drop and loading evolution in both cases. The model suggests that the coating has insignificant effect on the mass collection efficiency at clean state (2.7%), which after 6h diverges to 3.5% for the coated and 5.1% for the bare monolith (cumulative values). The final deposit loading value reveals that 58% of the amount collected in the channel is oxidized. This is attributed to the loose contact mechanism included in the model, while insignificant oxidation with NO_2 is expected (20% O_2, $4ppm$ NO_x). In contrast, the amount of deposit is collected at the leading edge does not change, implying absence of any catalytic effect in this region.

6 Outlook and conclusions

Loading experiments were conducted on bare and coated flow-through monoliths to study the effect of coating on particle collection mechanisms. To this end a model for bare monoliths was extended with a loose-contact oxidation mechanism to cover coated monoliths. The analysis of the experimental results using the model, showed that the coating has insignificant effect on the collection efficiency at clean state, however an appreciable effect is evident during the loading. 87% less mass is collected in the channel as a result of lower collection efficiency and catalytic oxidation of soot, while the buildup of deposit at the leading edge is not affected by catalytic coating. As a result, the coating induces a significant initial backpressure penalty accompanied by minimal increase during loading.

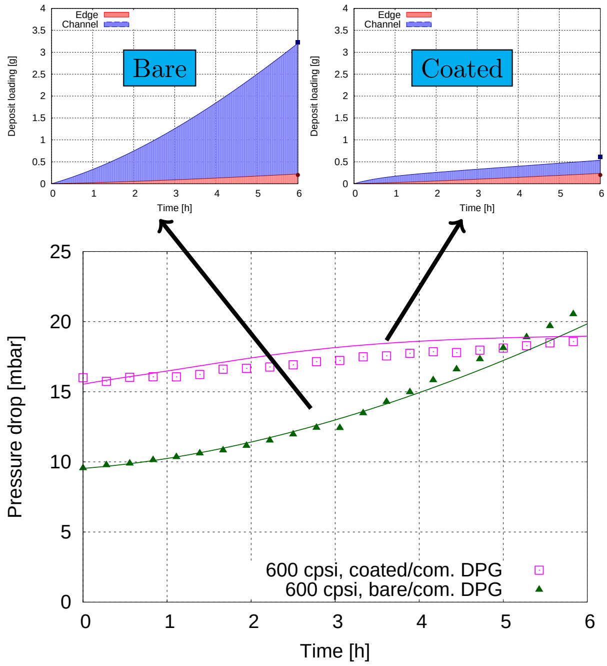


Figure 5: Effect of catalytic coating on monolith backpressure and deposit mass evolution.

References

[1] Payne S. D. Experimental studies of diesel particle filtration, PhD Dissertation, *University of Cambridge*, UK, **2011**

[2] Haralampous O., Payne S., Experimental Testing and Mathematical Modelling of Diesel Particle Collection in Flow-Through Monoliths, *Journal of Engine Research*, DOI: 10.1177/1468087416637735, **2016**

[3] Zeng L., Weber A. P., Transportmechanismen für aktivierten Sauerstoff in der katalytischen Rußoxidation an Nanopartikel-Schichtsystemen, *Chemie Inginieur Technik*, **2012**, 84, No. 3, 295-300