Washcoating and microstructure characterization of catalytic filters for exhaust gas aftertreatment

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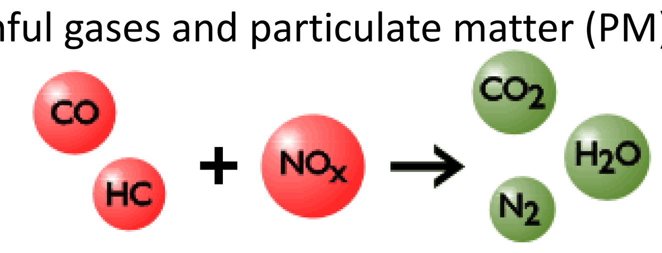
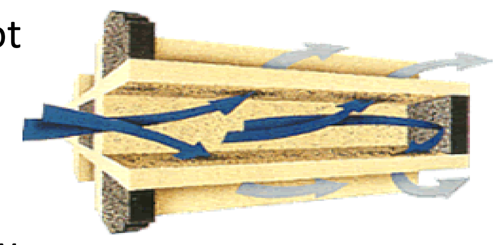
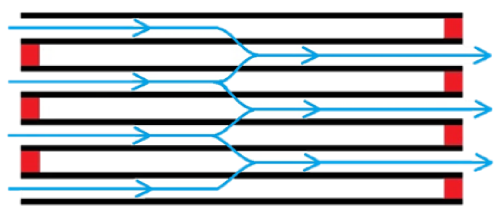
Introduction

- Combustion engines produce traces of harmful gases and particulate matter (PM)
- Gaseous emissions: CO, HC, NO_x
- Particulate matter: soot and ash
- **Catalytic particulate filter**
  - Removal gaseous emissions and filtering out the soot
  - Cylindrical monolith
  - Large number of parallel channels - honeycomb
  - Channels plugged alternately at one end ⇒ wall flow
  - Active layer (washcoat): γ-Al_2O_3 support with dispersed metals Pt, Pd, Rh, Ce
- Catalyst layer location: on/inside the wall – affects filter performance
  - **Pressure loss**
  - **Catalytic conversion**
  - **Filtration efficiency**

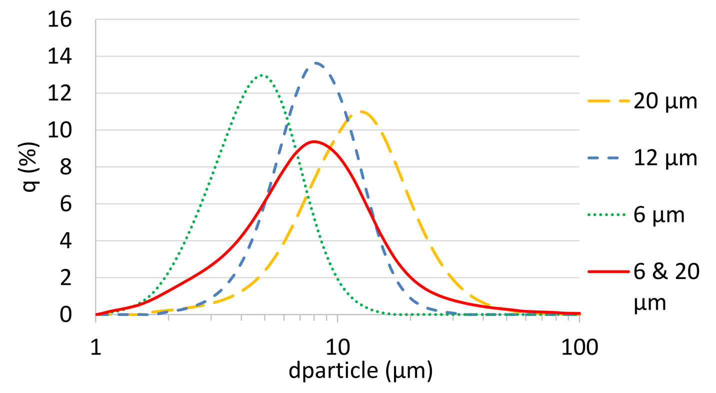






Preparation of porous layers

- Bare filter samples: cordierite ($2MgO{\cdot}2Al_2O_3{\cdot}5SiO_2$) monolith, d = 13 mm, L = 20 mm.
- The chosen method, used for thin film application ➡ **vacuum-deposition** *(-0.5 bar)*
- Coating material - slurry of Pt/γ-Al_2O_3
- Aqueous suspension 38 %
- The thickness and catalyst layer location depends on:
  - **Particle size d_{90}:** 6, 12, 20, 6 & 20 µm
  - **Amount of slurry:** approx. 1 g



Washcoating studied by time-resolved XRT

- Time-resolved XRT [1] at the TOMCAT beamline (Swiss Light Source)
  - Two heat guns (LE MINI from Leister) – 90°C
  - Modified rotary uninon (JR 1-1-4 R40 from TDS Precision Products GmbH)
  - Design allows transport of liquid between rotating and static parts
  - scanned volume 3188 × 3188 × 1600 µm³ , acquisition time 5s

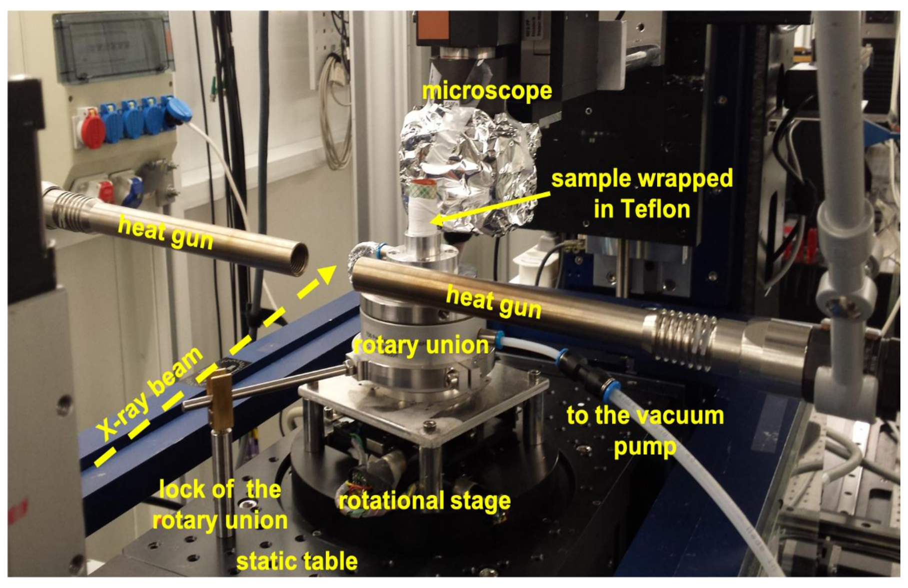


Fig. 1. Washcoating setup at the TOMCAT beamline (XRT) .

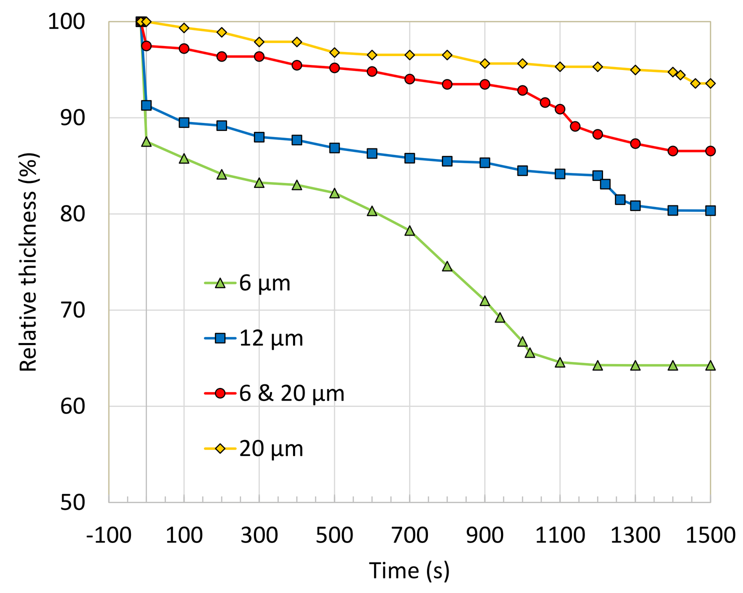
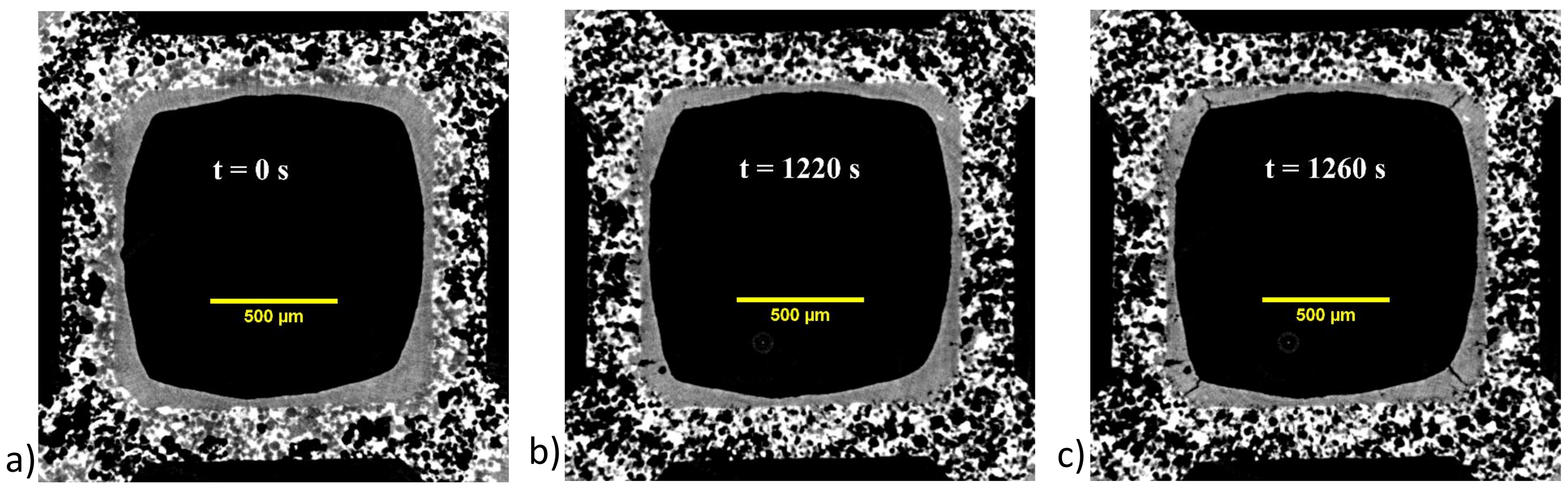


Fig. 2. Shrinking of the coated layer during drying.



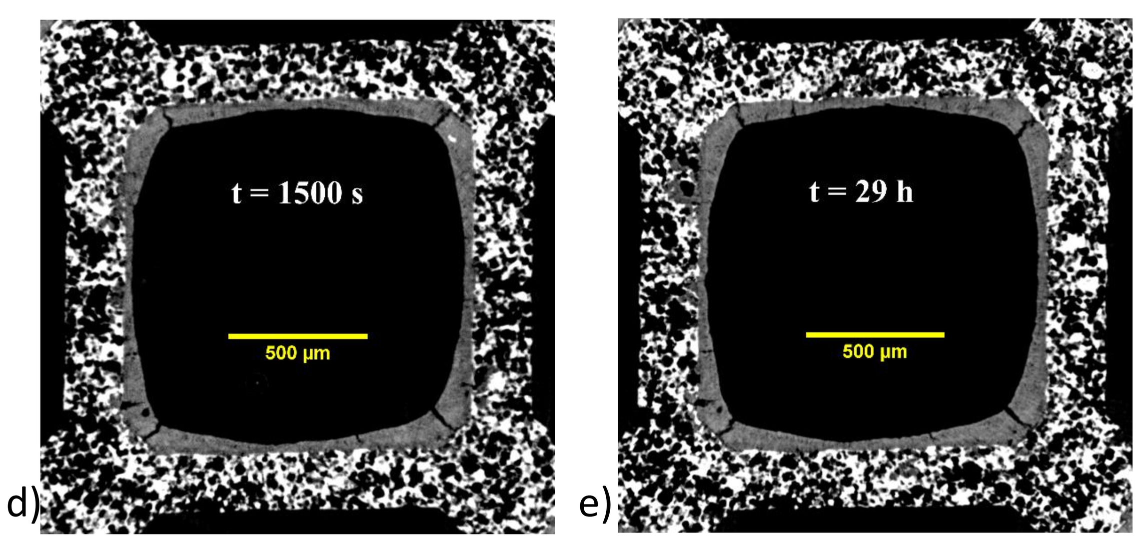
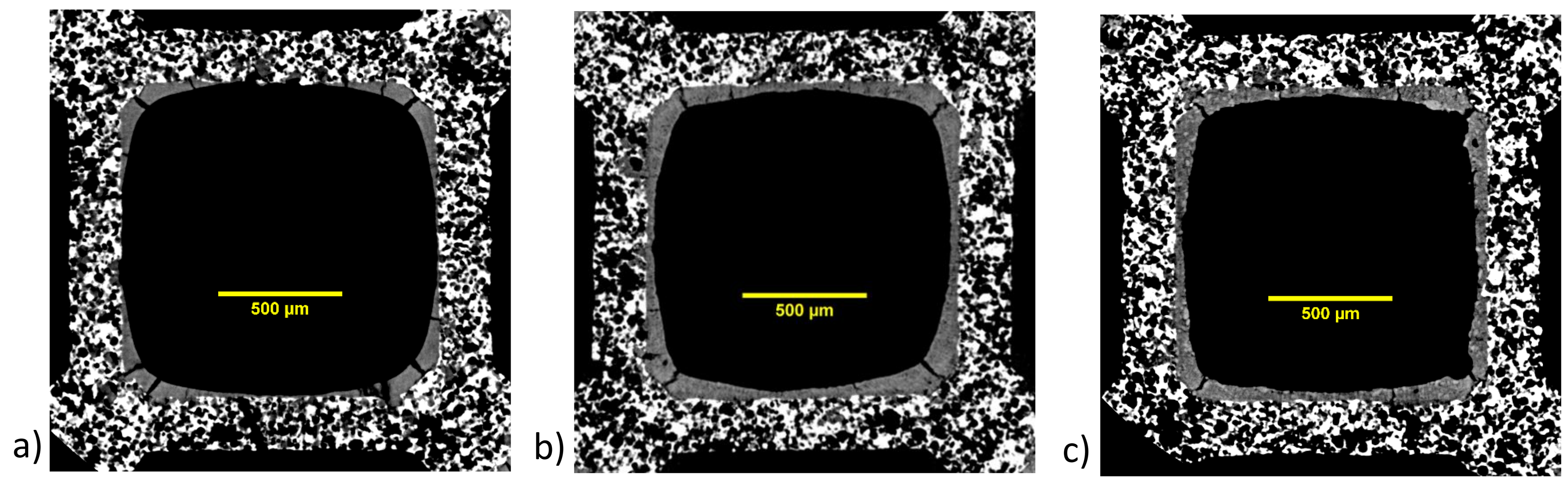


Fig. 3. Washcoat drying in a filter channel observed by XRT, sample d_{90} = 12 µm:
a) substrate after washcoating with the catalyst slurry (t = 0 s);
b) large pores inside the filter walls are dried, however the on-wall layer remains wet (t = 1220 s);
c) the walls are further dried, on-wall layer starts to shrink due to de-wetting, fcracks appear (t = 1260 s);
d) on-wall layer is dry (t = 1500 s);
e) final scan with a higher resolution (t = 29 h). White = substrate, grey = washcoat (brightness increases with water content), black = void

Final structures



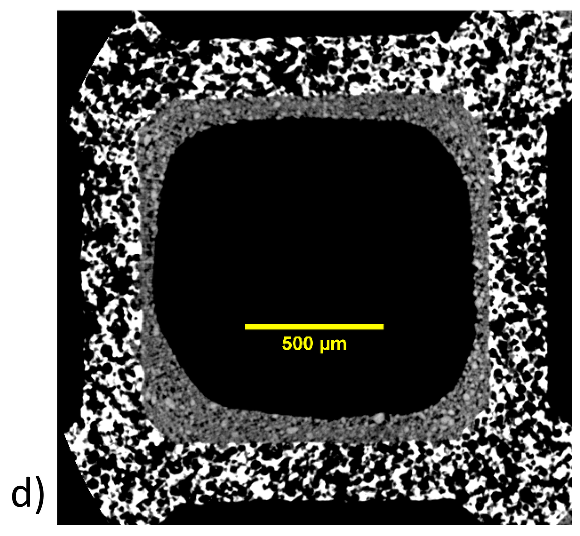


Fig. 4. Final structures after drying observed by XRT: a) d_{90} = 6 µm, b) d_{90} = 12 µm, c) d_{90} = 6 & 20 µm, d) d_{90} = 20 µm. White = substrate, grey = washcoat, black = void

Pressure drop

- The impact of coating on filter pressure drop:
  - Highest Δp: 6 & 20 µm – lower porosity, no cracks
  - Lowest Δp: 6 µm – thin layer, cracks

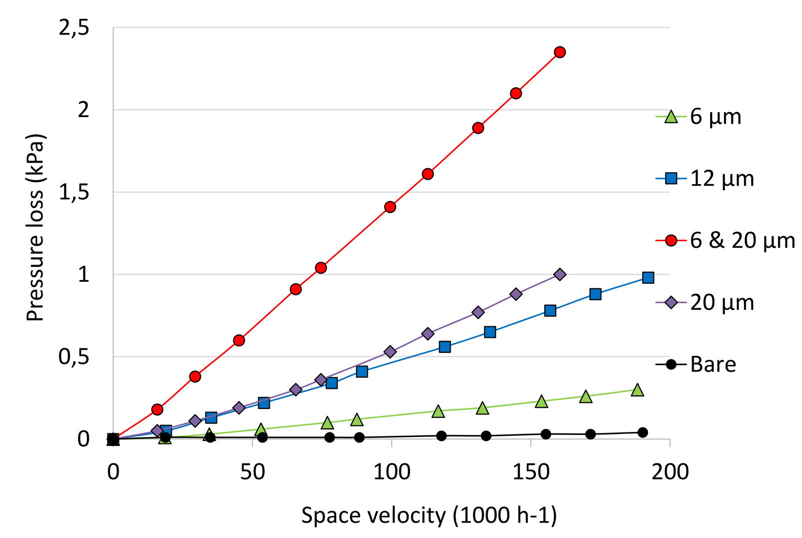


Fig. 5. Pressure loss in the coated filter samples depending on gas space velocity

Pore size distribution

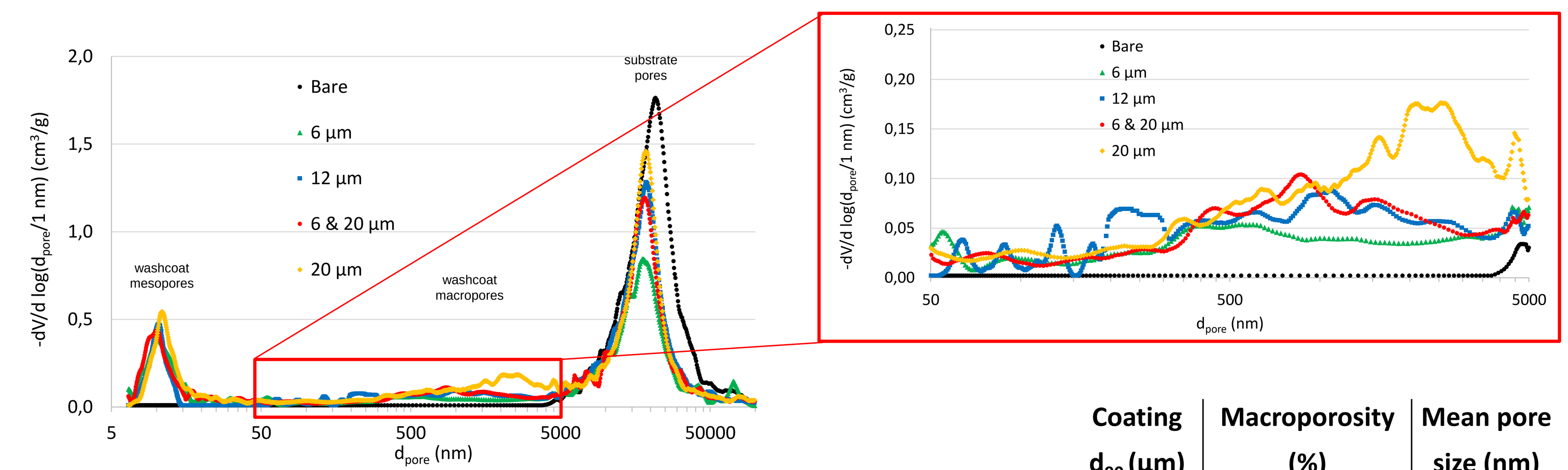


Fig. 6. Pore size distribution of bare and coated filter samples obtained by MIP: a) complete range, b) detail of washcoat macropore range

Coating d_{90} (µm)	Macroporosity (%)	Mean pore size (nm)
6	19.4	1274
12	26.7	1399
6 & 20	26.0	1347
20	37.6	1856

- Macropores in the substrate: maximum around 20 µm
- Macropores in the washcoat: maximum around 1-2 µm
- Mesopores in the washcoat: maximum around 10 nm

Conclusions

- The layer formed from the smallest particles (d_{90} = 6 µm) showed the highest tendency to crack, the highest in-wall coating fraction (approx. 60% of the catalyst inside the wall), and the lowest pressure drop.
- Slurry with particles d_{90} = 12 µm and larger, the coating was not able to penetrate the wall.
- The highest internal macroporosity and the largest macropores (beneficial for permeability) were achieved with the d_{90} = 20 µm coating, however, the thickest layer was formed on the wall, resulting in a high pressure drop.
- The processed XRT images can be further exploited for the generation of computational meshes and subsequent pore-scale simulations of permeation, diffusion, reaction and filtration in the reconstructed catalytic filters [2].

References

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