

The contact between platinum and soot particles in the catalytic oxidation of soot deposits on a diesel particle filter

Soot particles emitted by diesel engines could be efficiently removed from the exhaust gas using diesel particle filters (DPF). The critical issue concerning DPFs is their regeneration. Soot deposition leads to an increase of the pressure drop across the filter, thus decreasing the fuel efficiency of the engine. To regenerate the filter, soot particles are oxidized into CO and CO₂. The major problem is that soot particles can only be oxidized at temperatures higher than 550°C to 600°C with the oxygen contained in the exhaust gas. Normally the exhaust gas temperature does not exceed 250°C to 350°C at normal driving conditions. A possible solution is to use an active system to increase the exhaust gas temperature or the soot temperature. But it implies additional energy costs, requires complex means of control, and can create a significant thermal stress for the filter element. Therefore the use of passive systems, which decrease the oxidation temperature of soot through the use of catalysts is the preferred way to regenerate DPFs.

Today, the most widely used catalytically promoted regeneration process available for passenger cars is based on fuel borne catalysts (FBC). Organometallic compounds of transition metals (e.g. cerium, iron) or noble metal (e.g. platinum) additives are mixed with the fuel, leading to the formation of metal or metal oxide catalyst doped soot during the combustion in the engine. This catalyst can lower the soot oxidation temperature from 600°C to 450°C inside the DPF, but it is not sufficient to continuously regenerate it. Regularly, if the pressure drop is too high, additional post-injections of diesel fuel are performed to increase the exhaust gas temperature and oxidize the soot. The inconvenience with the FBC approach is the inherent requirement of providing a continuous additive supply in the fuel, which also leads to its continuous consumption and to the production and accumulation of “ash” inside the DPF, which decrease its life-time.

Research is now oriented towards alternative processes to regenerate DPFs in passenger cars. To avoid the ash accumulation and achieve a continuous regeneration, the idea is to apply the catalyst directly onto the filter. However, an effective way of oxidizing soot at reduced temperatures with a catalyst “coated” DPF has not been found yet, despite intensive technology development in recent years. The problem is related to the nature of the “contact” between soot and catalyst particles on the DPF, which seems to have a very strong influence on catalyst effectiveness in reducing the oxidation temperature.

The aim of this work was thus to determine if it is possible to achieve the same soot oxidation temperature reduction on a coated filter as the one obtain using a FBC. To compare the coated filter with the FBC technology, a unique catalyst had to be used. Platinum was chosen as the reference catalyst for the noble metal family. Three model contact configurations representing three different catalyst-to-soot contact scenarios on a sintered metal filter (SMF) were investigated.

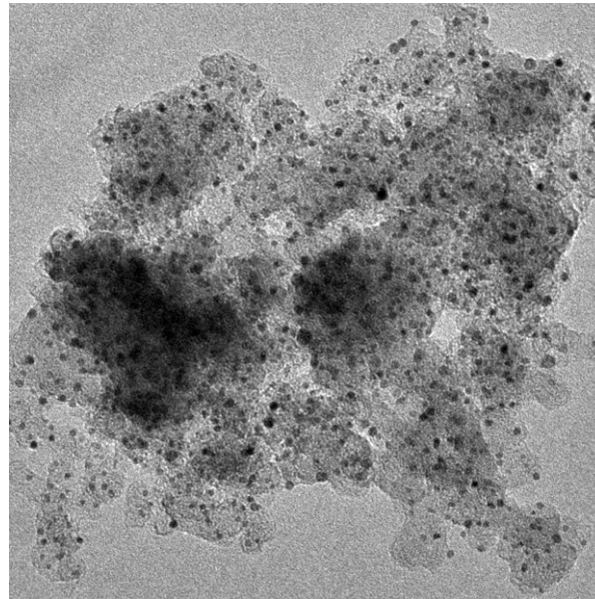
The first scenario represents the type of contact obtained using a fuel borne catalyst, which is known to be effective. It was prepared by filtering a platinum doped soot aerosol on the SMF. The second scenario represents a typical loose contact obtained on conventional coated filters. It was achieved by first filtering a platinum aerosol, and then the soot aerosol. In the third scenario, platinum and soot aerosol were filtered simultaneously, representing a uniform distribution of platinum in the soot cake. Temperature programmed oxidation (TPO)

experiments were performed in a specially designed reactor to determine the effectiveness of each configuration on the basis of its ability to lower the soot oxidation temperature. Experiments were performed using two different soot sources (produced with a spark discharge generator or by pyrolyzing a toluene aerosol), that exhibit the same results.

It was found that platinum doped soot deposition on the filter has the same catalytic oxidation effect as mixing platinum and soot particles in the filter cake by simultaneous filtration. For simultaneous filtration the soot oxidation temperature was reduced by up to 140°C to 250°C for platinum quantities varying between 3 and 47 %-wt in soot cake. On the other hand, the consecutive deposition of a platinum and a soot layer produced only a slight temperature reduction in the order of about 15 to 25°C. These results show that the platinum catalyst is as effective on a filter as it is in the aerosol state in reducing the soot oxidation temperature, provided platinum particles are distributed uniformly throughout the soot cake. Apparently, the distance between soot and platinum particles plays a key role in promoting an effective oxidation on the filter, which is consistent with the role of platinum particles as local generators of activated oxygen.

To conclude, it is possible to achieve the same soot oxidation temperature decrease on a coated filter as the one obtained using a FBC, if the platinum particles are uniformly distributed in the soot cake. The challenge in designing effective coated filters is thus to develop new coatings allowing a depth filtration of the soot, so that it can increase the contact surface between platinum and soot particles greatly.

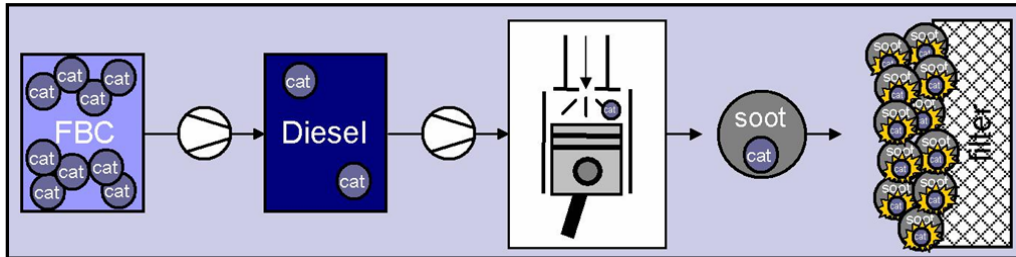
On the effect of the contact between platinum and soot particles on the catalytic oxidation of soot deposits on a diesel particle filter



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| K. Hinot | Deutsches Zentrum für Luft- und Raumfahrt e.V. (DLR), Germany |
| H. Burtscher | IAST, University of Applied Sciences, Switzerland |
| A. Weber | IMV, Technische Universität Clausthal, Germany |
| G. Kasper | IMVM, Universität Karlsruhe, Germany |

Motivation

Fuel borne catalyst (FBC):

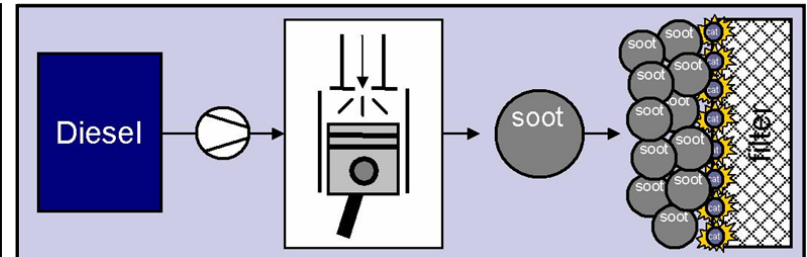


Soot oxidation temperature decreases to ca. 450°C



Catalyst consumption
accumulation of ashes

Coated filter:



No reduction of the soot oxidation temperature



One-time catalyst coating

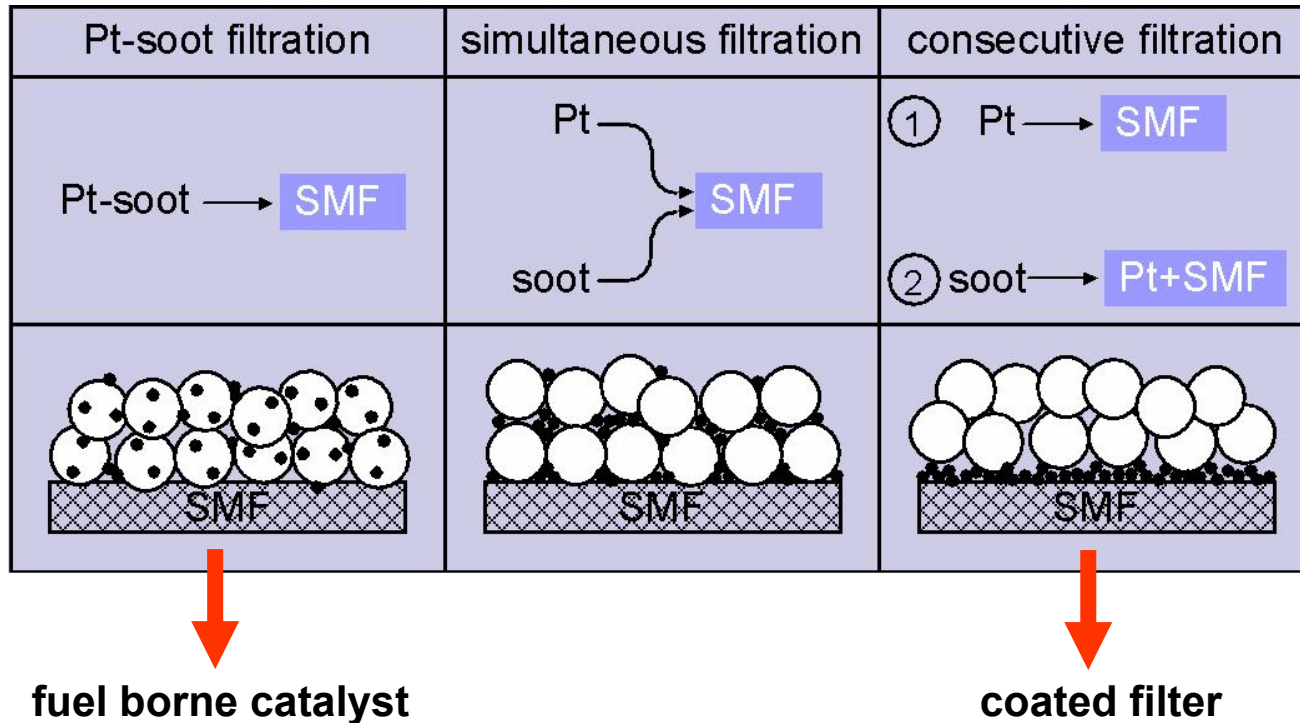
- **Is it possible to achieve the same soot oxidation temperature decrease on a coated filter as with a FBC?**

→ choice of a catalyst: **Platinum**

Methodology

■ 3 different platinum and soot aerosol filtration scenarios:

- on sintered metal filter (SMF) sheets
- with two model soot aerosols



Soot generated by a spark discharge generator

■ SDG-soot aerosol:

→ 7 nm particles

■ Pt aerosol:

□ SDG (2 Pt electrodes)

→ 3 nm Pt-particles

■ Pt-doped SDG-soot aerosol:

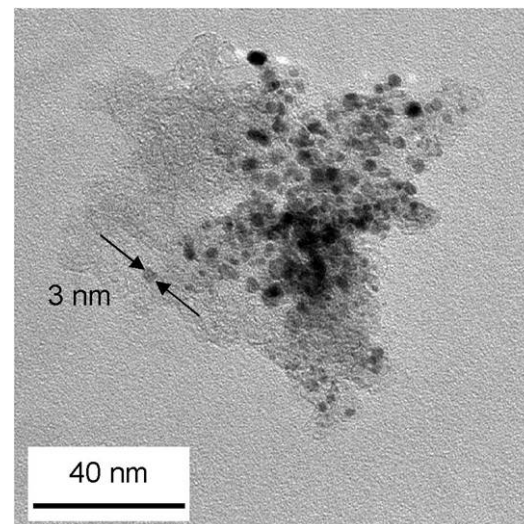
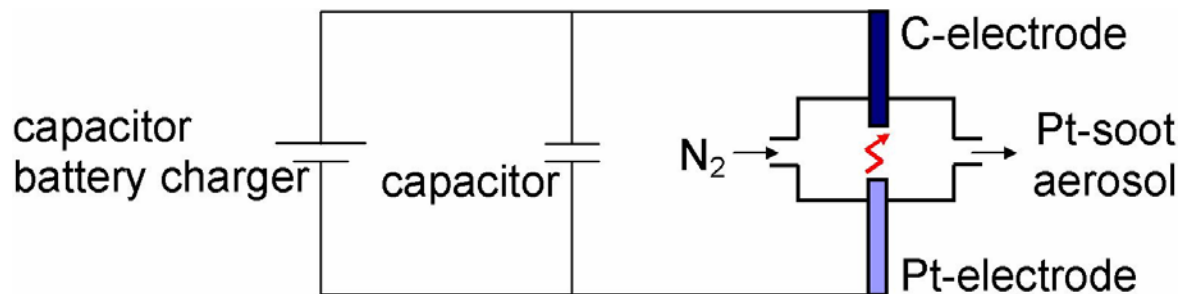
□ SDG (1 Pt – 1 C electrode)

→ 47 %-wt Pt

→ 3 nm Pt-particles

→ 7 nm soot-particles

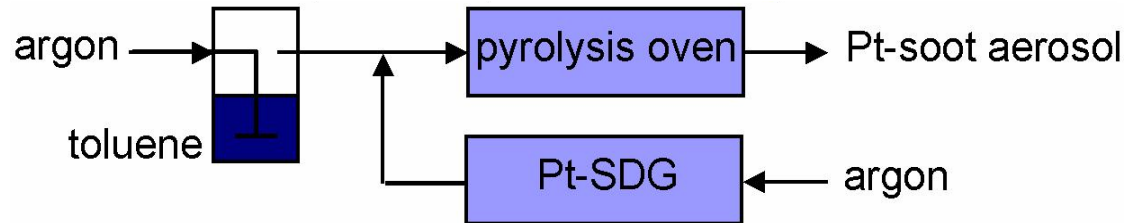
→ Pt on soot



Soot generated by pyrolyzing a toluene aerosol

■ Pyrolysis-soot aerosol:

→ 16 nm particles



■ Pt aerosol:

□ SDG (2 Pt electrodes)

→ 3 nm Pt-particles

■ Pt-doped pyrolysis-soot aerosol:

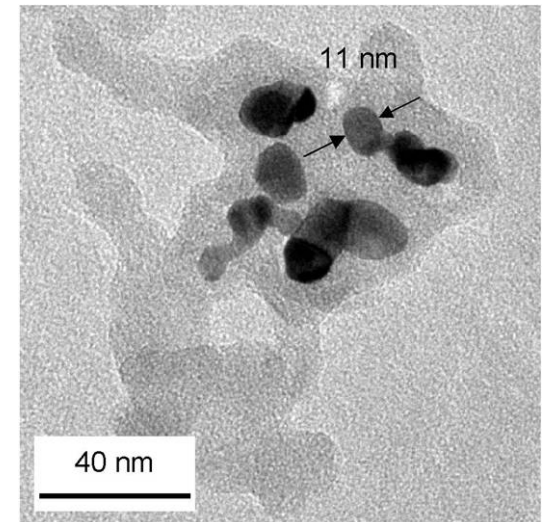
□ Toluene droplets diluted in a Pt aerosol (produced by SDG)

→ 3 %-wt Pt

→ 16 nm soot-particles

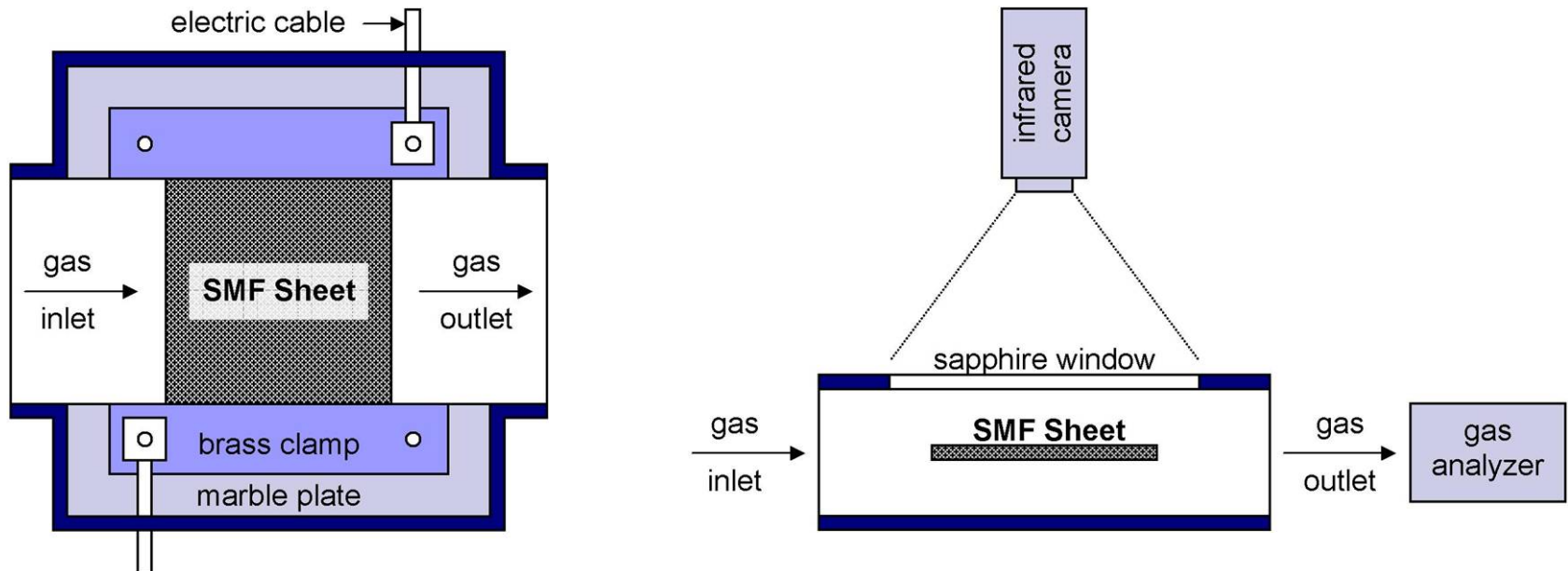
→ 11 nm Pt-particles

→ Pt in soot



Deposited soot oxidation experiments

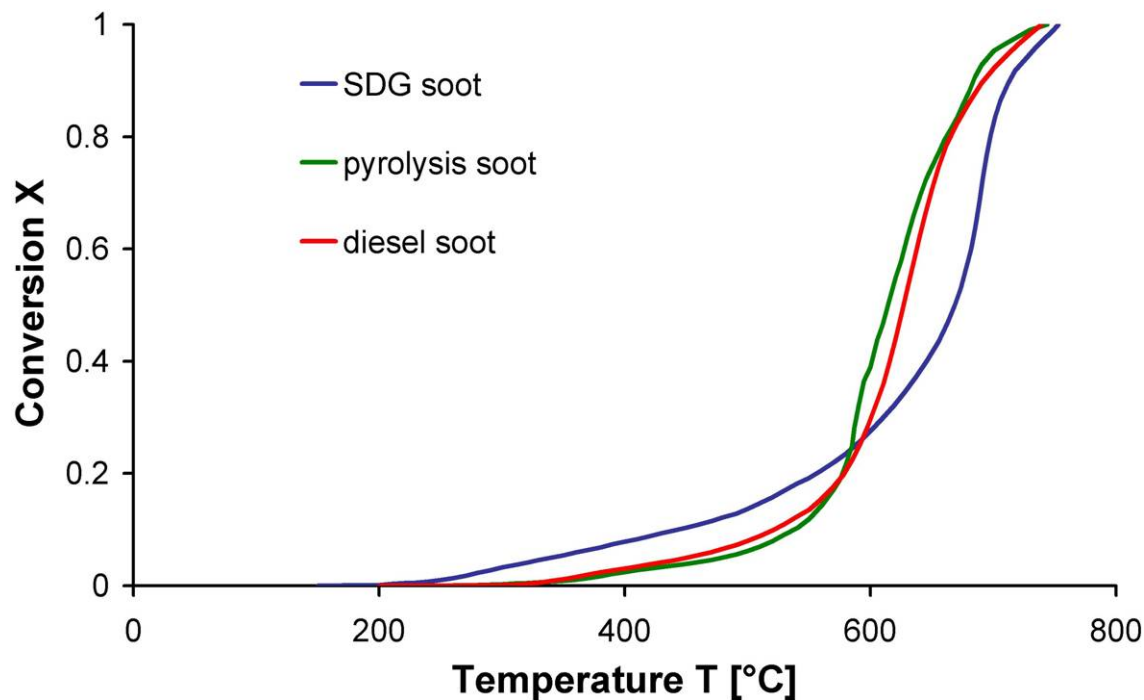
■ Reactor and experimental set-up:



■ Temperature programmed oxidation:

- 10 K/min, 10 % O₂ in N₂
- CO/CO₂ concentration → X
- soot temperature with IR-camera → T

Thermal soot oxidation

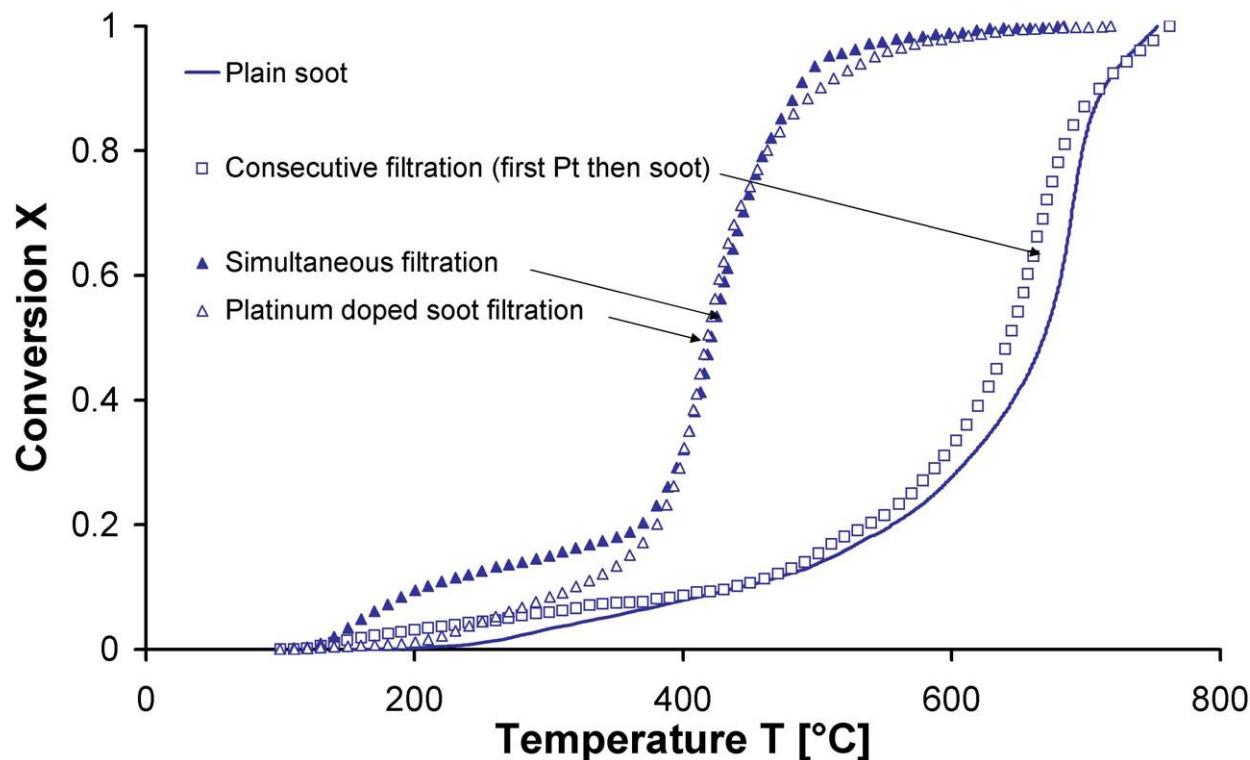


	C (%-wt)	H (%-wt)	N (%-wt)	O (%-wt)	d_{SMPS} (nm)	d_{TEM} (nm)	S_{BET} (m ² /g)
SDG soot	86.7	0.6	8.0	4.5	70	7	700
pyrolysis soot	96.5	1.4	<0.1	1.9	110	16	140
diesel soot	90.9	1.4	0.9	6.6	110	18	160

Influence of the filtration scenario (1)

■ SDG-soot with 47 %-wt platinum:

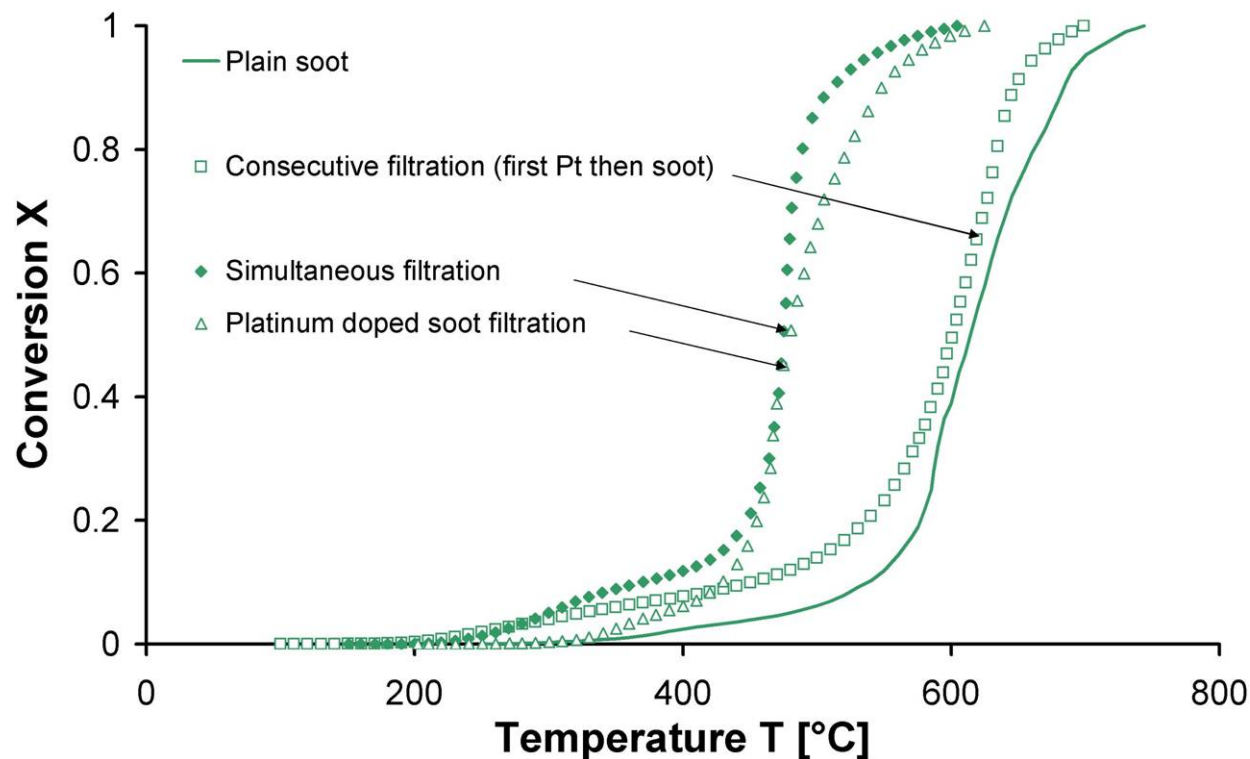
Filtration	$T_{0.5}$ (°C)
Plain soot	669
Consecutive	643
Simultaneous	421
Pt-doped soot	418



Influence of the filtration scenario (2)

■ Pyrolysis-soot with 3 %-wt platinum:

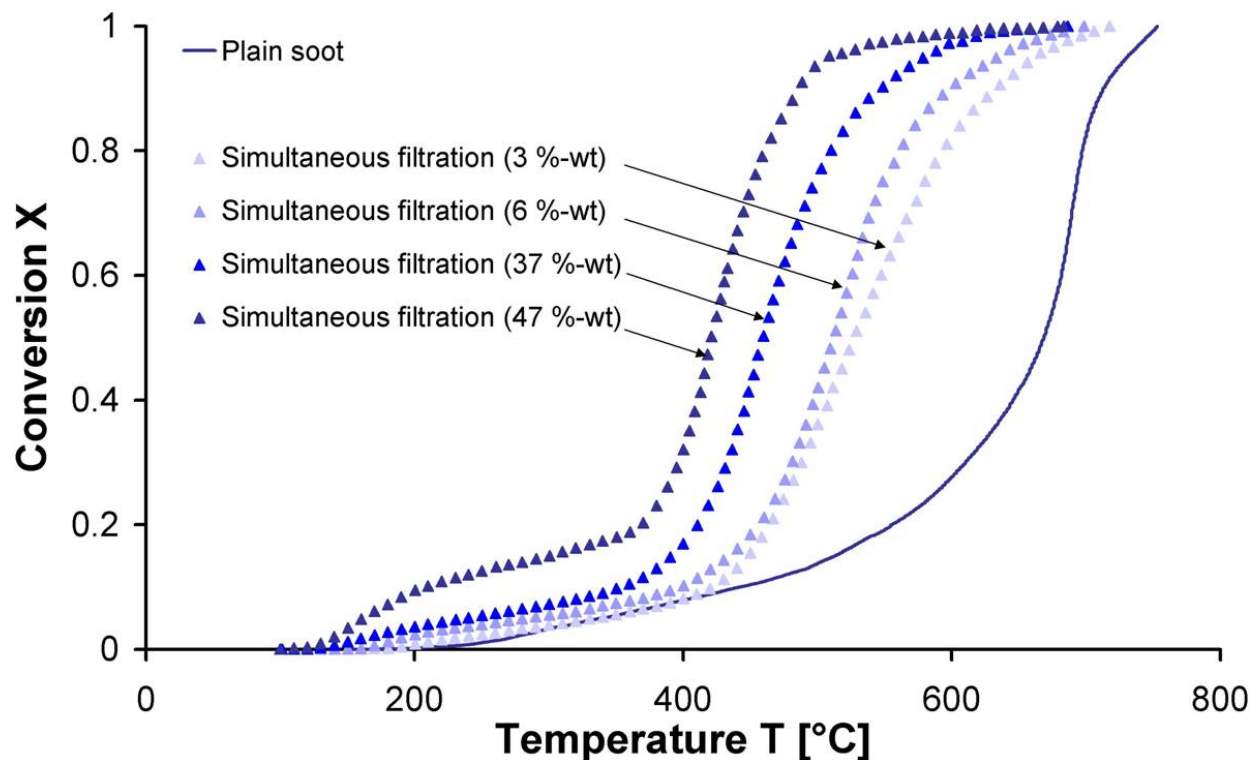
Filtration	$T_{0.5}$ (°C)
Plain soot	614
Consecutive	601
Simultaneous	475
Pt-doped soot	479



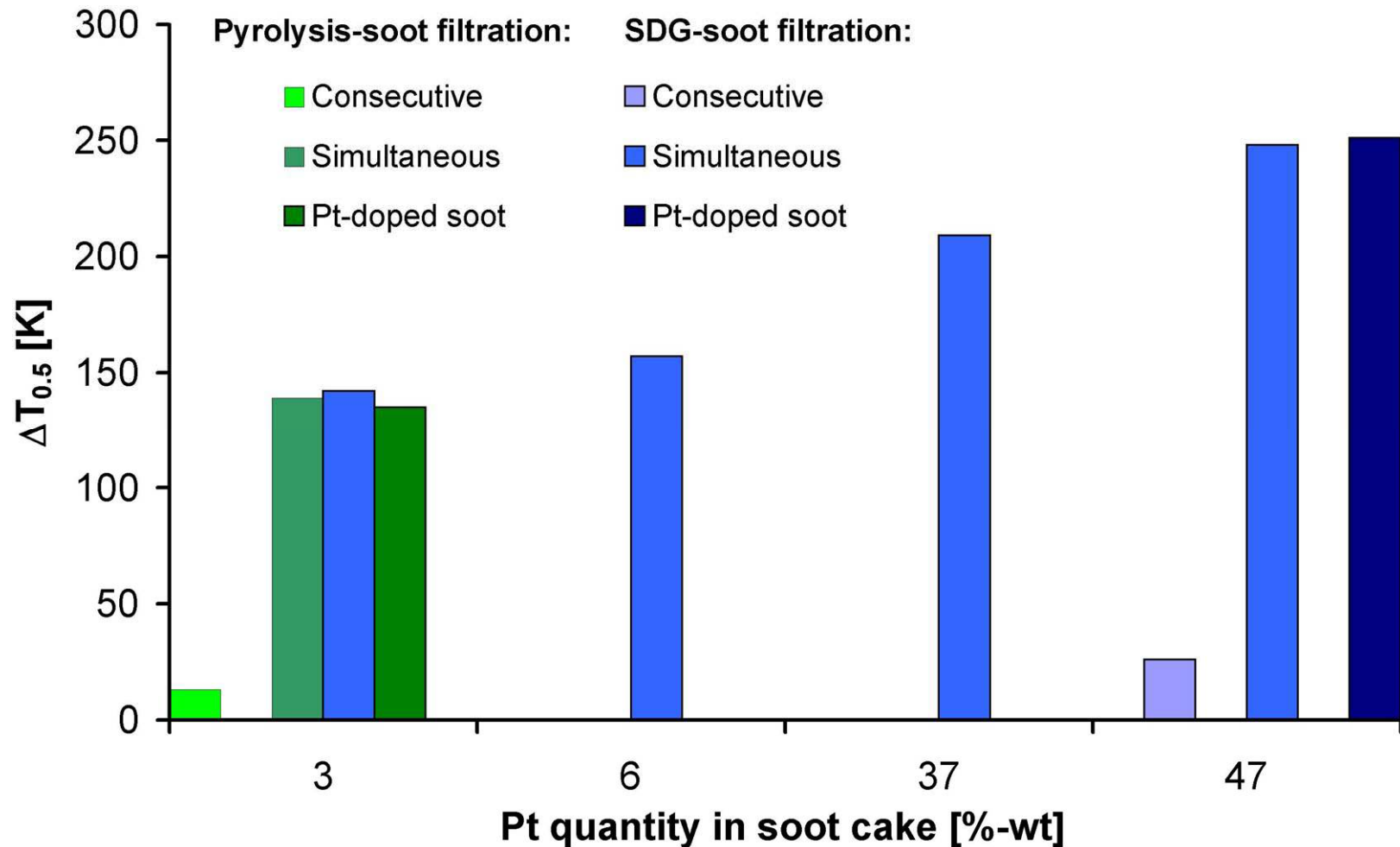
Influence of the Pt-quantity in soot cake

■ Simultaneous filtration (SDG-soot):

Pt (%-wt)	$T_{0.5}$ (°C)
0	669
3	527
6	512
37	460
47	421



Comparison



Results

■ Thermal soot oxidation:

- **Pyrolysis-soot aerosol** presents the closest structure/composition **and** the closest conversion curve to that of reference **diesel-soot aerosol**
- **SDG and pyrolysis soot have similar soot oxidation behavior than diesel soot**

■ Catalytic soot oxidation:

- $\Delta T_{0.5}$: **Pt-doped soot filtration \approx simultaneous filtration \gg consecutive filtration**
independent of: soot aerosol (SDG or pyrolysis)
 Pt-quantity in soot cake (47 or 3 %-wt)
- **Simultaneous filtration: $\Delta T_{0.5}$ increases with the Pt quantity in soot cake**
- **effectiveness: only if Pt particles distributed uniformly through the soot cake**

Conclusions

- **Is it possible to achieve the same soot oxidation temperature decrease on a coated filter as with a FBC?**
 - **YES**, if the **platinum** particles are **uniformly distributed** in soot cake
 - The method can be used for other catalyst (metal oxide family)

- **Further research field:**
 - Design of effective coatings allowing a **higher density** of catalyst in soot cake
 - **Depth filtration coatings**



**Thank you
for your attention!**