Development of Functionally Graded SiC-Based Diesel Engine Exhaust Gas Filter

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ABSTRACT

The development of a SiC-carbide based filter that combines a low pressure drop due to a graded pore structure with high filtration performance ascribed to different filtration mechanism is presented.

The processing route is based on carbon paper/carbon black composites with graded porosity. By microwave assisted chemical vapor infiltration (CVI) the composite is coated with silicon and partially carbidized in a way, that the oxidation resistivity of the obtained SiC is combined with the good mechanical strength of the remaining carbon fiber core.

To reduce the soot removal temperature during operation, the material is coated with a Platinum/TiO_2-catalyst.

Keywords: particle filter, diesel soot, microwave, chemical vapour infiltration

1 INTRODUCTION

The incomplete combustion of diesel fuel in diesel engines as well as the recombination in the exhaust gas leads to carbon particle, inorganic oxide, and hydrocarbon emissions. The particle size varies between 5–20 nm up to 50 – 150 nm for agglomerates [1].

Because of the extreme pressure drop filter material with bulk pore sizes in the range of the soot particles are not useful. Therefore the filter materials consist of large pores and the particles are collected by “Deep-bed-filtration” (impaction and diffusion) and “Cake-filtration” instead of “Sieve-filtration”.

Hot gas filtration using ceramic filters for diesel engine exhaust gas cleaning has gained considerable attention and growth over the last 15 years now.

State of the art diesel particle filters (DPF) utilize mainly cordierite or silicon carbide wall-flow monoliths with a honeycomb structure to trap the soot produced by diesel engines. Exhaust gases escape through the pores in the wall material and most of the soot particles are trapped in the filter walls.

The requirements of the removal efficiency, especially for extreme fine particles, leads to a high pressure drop in the filter cartridge and a significant increase of fuel consumption.

Therefore the development goal was a SiC-carbide based filter that combines a low pressure drop due to a graded pore structure with high filtration performance ascribed to different filtration mechanism.

To achieve the different requirement of a modern DPF, different performance parameters has to be taken into account (see figure 1). The filter wall material development has to consider conflictive requirements like high permeability and high particle collection efficiency.

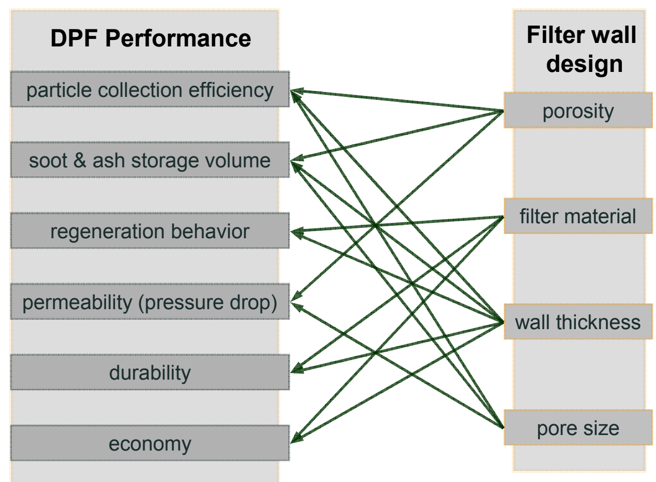

Figure 1: Diesel particle filter (DPF) development.

During exhaust gas filtration from time to time it is necessary to clean the DPF by a thermal treatment, because the collected particles enhance pressure drop.

Soot particle start burning at temperatures higher than 600°C but the burn off can be promoted by catalysts down to temperatures of 300–400°C [2].

2 EXPERIMENTAL

An overview of the DPF processing steps are shown in figure 2.

The material processing starts from thin commercial carbon papers with low pressure drop but insufficient removal efficiency for nano-scale soot particles. To achieve a high particle removal without a strong increase of pressure drop, from one side a carbon black slurry is partially infiltrated into the carbon paper. At this stage the composite can be folded into complex shapes to increase the filtration area.

To achieve the necessary oxidation resistivity at high temperatures, the material is heated in a 2.45 GHz microwave cavity while trichlorosilane is poured though the porous material (see figure 3).

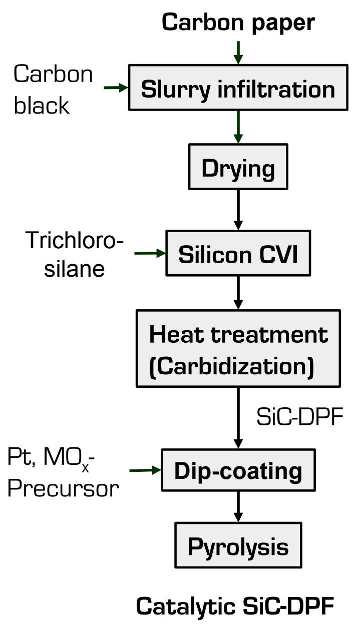

Figure 2: Diesel particle filters (DPF)-material preparation process

The thermal decomposition of the trichlorosilane can take place either homogeneous in the gas phase followed by an impaction and/or diffusion of the nano scale silicon particles at the carbon surface or by heterogeneous decomposition of the precursor at the carbon surface (chemical-vapour-deposition). The simplified decomposition reaction is given by:

$$SiHCl_3 + H_2 \leftrightarrow Si + 3\ HCl$$

During an additional heat treatment at 1500°C the temperature stable SiC is formed.

In the next processing step a Platinum/TiO2 –containing wash-coat is infiltrated into the DPF-material, to reduce the soot burn off temperature during the exhaust gas filtration. A detailed description of the coating procedure is given in [4].

The flow rate of a filter is dependent on the applied differential pressure and is one of the most important parameters to characterize a DPF.

The non linear relation between pressure drop and flow rate is described by the following equation:

$$\Delta p = \frac{\dot{V} \cdot s}{A} \cdot \left[\frac{\eta}{\alpha} + \frac{\rho \cdot \dot{V}}{\beta \cdot A} \right]$$

ρ = Viscosity coefficient [m^2]	*β = Inertia coefficient [m]*
s = Filter thickness [m]	*$\dot{V}$ = Flow rate [m^3/s]*
ρ = Fluid density [kg/m3	*A = Filter surface [m^2]*
ρ = Dynamic viscosity [Pa•s]	*$\Delta\rho$ = Pressure drop [Pa]*

The equation can be divided into a laminar (linear) and a turbulent part.

For laminar flow, the simplified Darcy's equations can be used to show the relation between the variables:

$$\Delta p = \frac{\dot{V} \cdot s \cdot \eta}{A\alpha}$$

The flow late measurements have been performed up to pressure drops much higher than during operation of the filter material in the exhaust gas system of diesel engines.

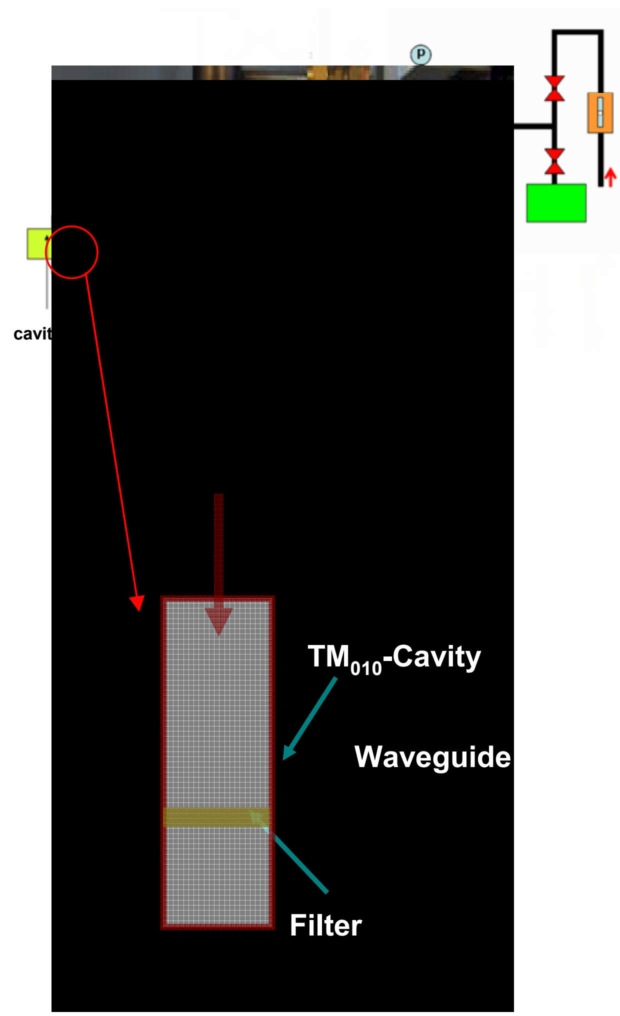

Figure 3: Experimental set-up for the microwave assisted trichlorosilane decomposition on the carbon pre-form.

3 RESULTS

The development of the microstructure during the different process steps is shown in figure 4.

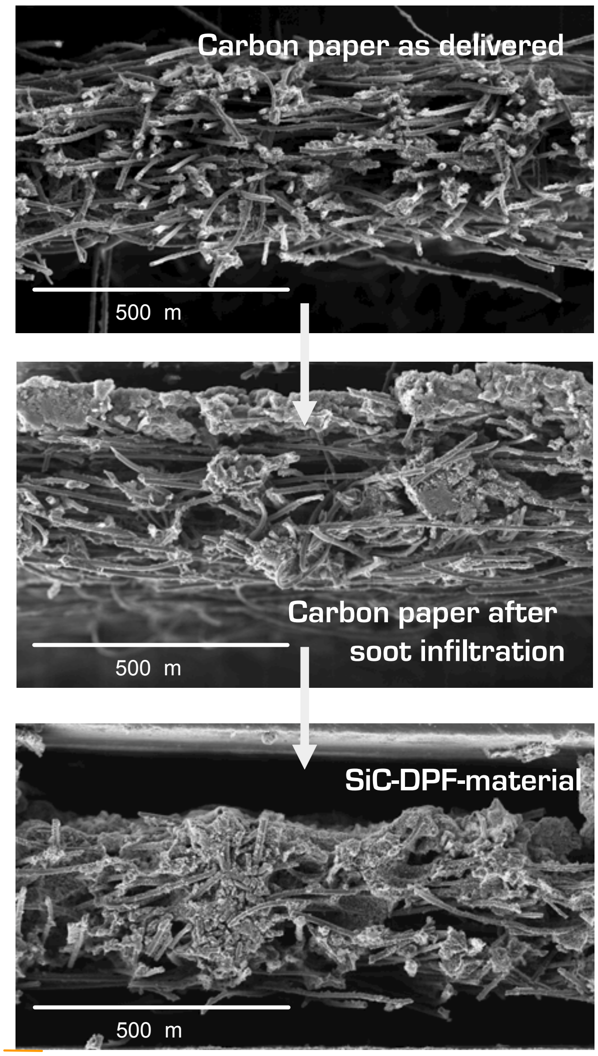

Figure 4: SEM-Cross section of the material after different processing steps

After dying, the carbon black infiltrated carbon paper material consists of graded porosity with a top layer of small pores and an increasing pore size down to large pores at the bottom.

By microwave decomposition of the trichlorosilane it is possible to cover the carbon black particles as well as the carbon fibres with fine grained silicon.

A SEM picture of the silicon coated carbon fibre is shown in figure 5.

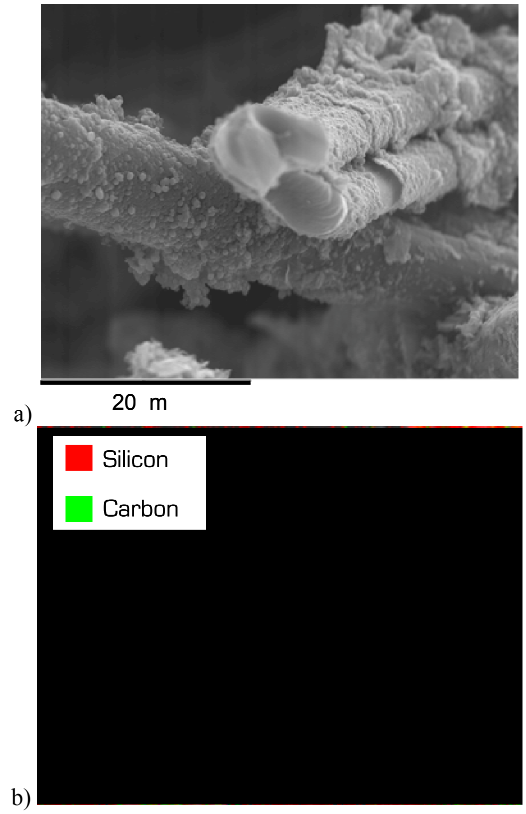

Figure 5: SEM picture of a silicon coated carbon fibre and EDX-analysis of the carbon and silicon distribution

During the following heat treatment the carbon black is converted to SiC with maintaining the pore structure while the carbon fibres are partial converted to utilize the good mechanical properties of the carbon-fibre.

The XRD-analysis of the material after carbidization is shown in figure 6. The remaining carbon core enables an easy handling like bending, cutting and assembling of the filter material.

The resulting pore size distribution of the DPF compared to the pore size distribution of the carbon paper is shown in figure 7.

While the carbon paper meanly contains pores in the range of 30 to 300 m, the DPF has additional pores in the range of 100 nm. Even if these small pores are located in the small top layer of the filter, a strong effect on the permeability of the filter material can be measured (see figure 8).

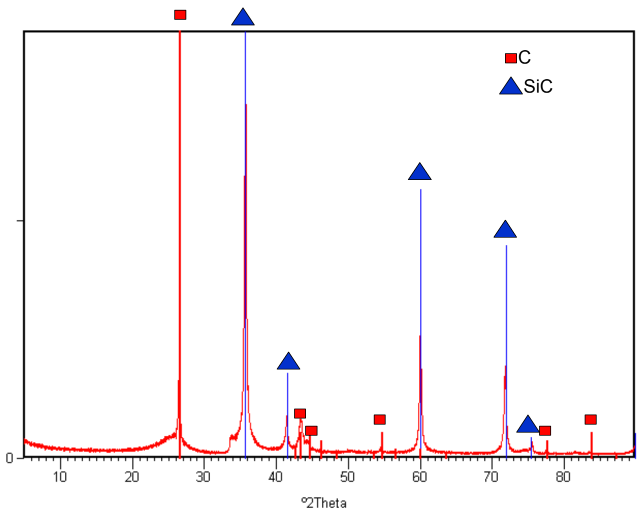

Figure 6: XRD-Phase analysis of the DPF after carbidization

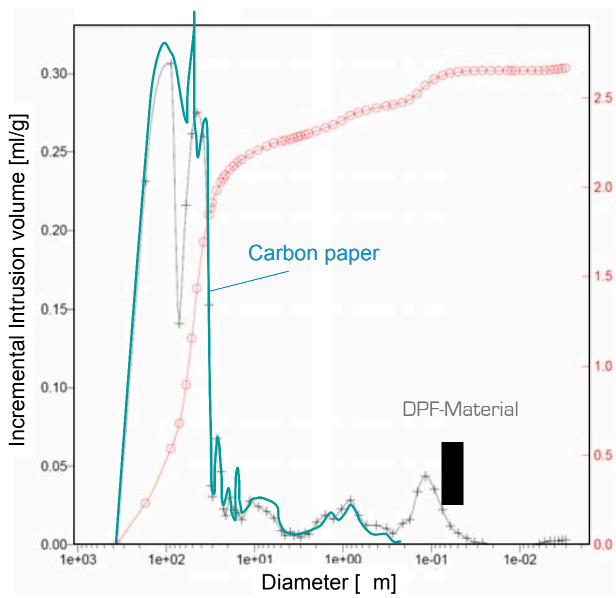

Figure 7: Pore size distribution of the DPF compared to the carbon paper as delivered.

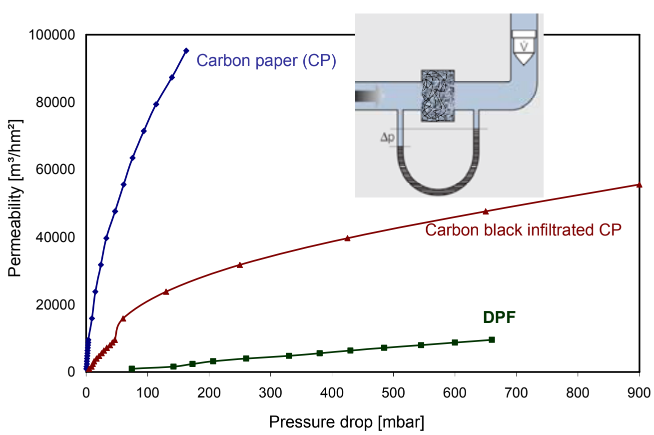

Figure 8: Permeabilty of the DPF compared to the carbon paper and the carbon black infiltrated carbon paper

4 CONCLUSION

A new processing route for a functionally graded deep bed DPF-system has been developed. Each step of the processing and also the selection of the material components (carbon paper, carbon black, standard catalysts trichlorosilane) enables an easy scale up.

Up to now the permeability of the DPF is in the same range as commercial filter systems, therefore especially the carbon black infiltration has to be further optimized to reduce the pressure drop in the top layer.

It could be shown that the microwave assisted pyrolysis of the silicon precursor (Trichlorosilane) offers an ideal method for homogeneous and controllable coating of the carbon material with silicon.

The carbon fiber core enables an easy handling and assembling of the filter.

Because of the graded pore size and porosity also the typical filter parameter like particle collection efficiency, storage volume and pressure drop are graded as shown in figure 8.

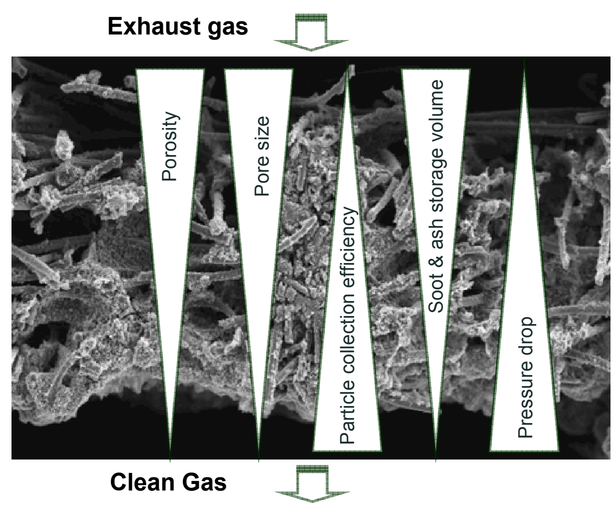

Figure 8: Functionally gradient concept for DPF

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