

# Particulate Matter Trapping and Oxidation on a Catalyst Membrane

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## ABSTRACT

Particulate matter (PM) trapping and oxidation in regeneration on the surface of a diesel particulate catalyst-membrane filter (DPMFs) were investigated in detail using an all-in-focus optical microscope. The DPMF consists of two-layer sintered filters, where a SiC-nanoparticle membrane (made from a mixture of 80 nm and 500 nm powders) covers the surface of a conventional SiC filter. Using a visualization experiment, it was shown that PMs were trapped homogeneously along fine surface pores of the membrane's top surface, whereas in the regeneration process, the particulates in contact with the membrane may have been oxidized with some catalytic effect of the SiC nanoparticles. A soot cake was reacted continuously on the nanoparticles since pushed by a gas flow. The oxidation temperature of particulate trapped on the SiC-nanoparticle membrane was about 75 degrees lower than that on the conventional diesel particulate filters (DPF) without a catalyst. The activation energy of reaction on the nanoparticle membrane became lower than that on the conventional SiC-DPF without catalyst. Consequently, the SiC-nanoparticles membrane filters had some catalytic activity similar to that of the conventional catalyst DPF.

## INTRODUCTION

Among internal combustion engines, diesel engines have the highest thermal efficiency for a given output power; however, particulate matter (PM) and NO<sub>x</sub> must be removed from exhaust gases emitted from diesel engines to protect the environment and human health. Creating an environment friendly power source for next-generation diesel engines requires high-efficiency emission control. Diesel particulate filters (DPFs) play an important role in particulate trapping and oxidation. Although these processes involve a complex behavior of particulates and reaction phenomena, little

information is available to aid in understanding such phenomena.

A number of studies have been performed to simulate particulate trapping and chemical reactions inside DPFs to provide valuable information in designing them [1,2,3,4,5,6]. However, optimization of the modern diesel engine combustion process has led to an increase in the production rate of nanosized particles. The nano and microstructures of soot particles were characterized with transmission electron microscopy (TEM) in order to understand them in detail [7,8]. Furthermore, the overall oxidation reaction of soot was investigated by performing temperature programmed oxidation [9]. Recently, catalytic fuel additives and catalyst DPFs were proven to reduce the overall activation energy required for particulate oxidation, resulting in lower-temperature oxidation compared to noncatalyst DPFs. However, complete contact was not necessarily expected between the soot and the catalyst particles on the DPF wall [10].

<figure 1 here>

Through macroscopic [11,12] and microscopic [13,14,15] visualization experiments, particulate trapping behavior and active regeneration inside a DPF were investigated successfully for better understanding and future design of an ideal DPF configuration. We noted that reducing emission at the beginning of trapping, where the filtration efficiency was not so high, was required, as shown in Fig.1. At the beginning of trapping, some diesel particulates with diameters of around 100 nm pass through the DPF wall microchannels of average size 11 μm because the inertia and the Brownian motion effects were small for trapping. After relatively large-scale particles were trapped around the bottom of the surface pore by the inertia effect, the filtration efficiency increased with time. Finally, after the soot cake has

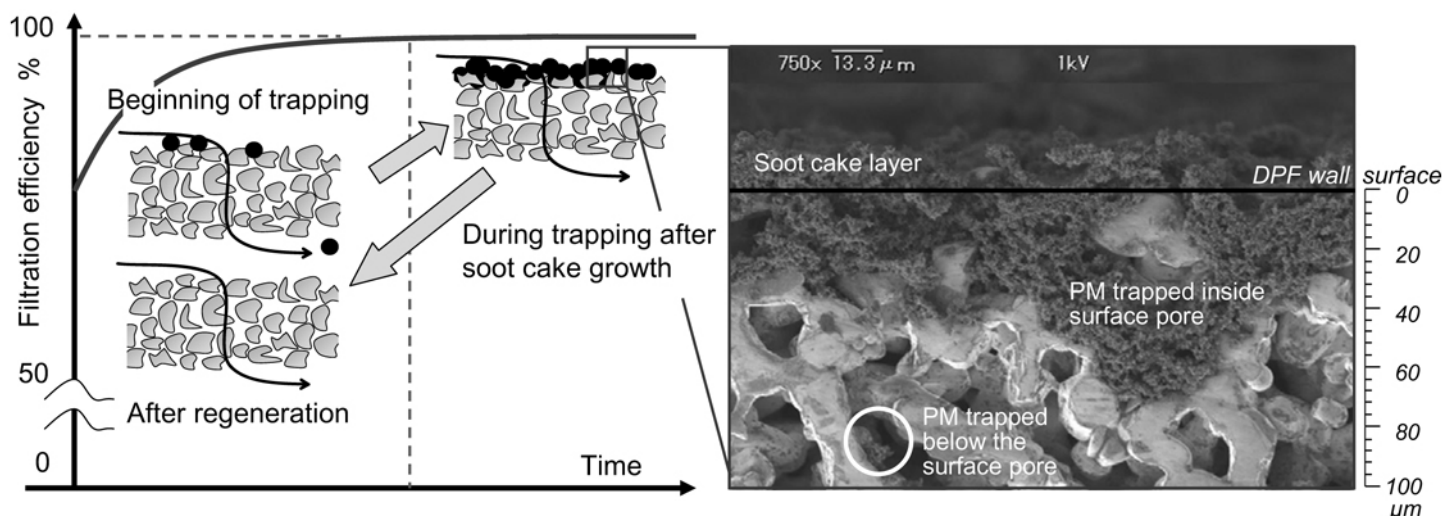


**Figure 1. Filtration efficiency of a conventional DPF and SEM image [15] of the particulates trapped inside DPF surface pores.**

grown, the efficiency became almost 100%. As shown in the SEM image [15] in Fig.1, the surface pores are filled with particulates, and only a few particulates, which may have passed through at the beginning of the trapping process, are trapped in the deeper pores. However, after the DPF is regenerated, the filtration efficiency decreases again.

In previous studies, a concept of an ideal DPF, in which a micron-sized particle layer was pasted and sintered on the DPF wall surface instead of the soot cake, was proposed to improve the filtration efficiency at the beginning of the trapping process [16,17]. Furthermore, a catalyst membrane filter (made of zeolite with sodium, potassium, or cesium) pasted onto the conventional SiC-DPF was also proposed and developed successfully [18,19,20]. Microchannels in the membrane filter were much smaller than that in the conventional SiC-DPF.

In the present study, a nanoparticle membrane filter was proposed on the basis of the previous studies. It consists of a SiC-nanoparticle membrane filter sintered onto the conventional SiC-DPF. The PM trapping and oxidation in regeneration were investigated through microscopic visualization experiments and gas analysis during regeneration.

## EXPERIMENTAL SETUP

### EXPERIMENTAL PROCEDURE

A diesel engine (Robin SDG2200, manufactured by Subaru Co. Ltd.) with a displacement of 230 cm<sup>3</sup> and a rated output of 2.8 kW was used in the trapping process. The fuel injection pressure was approximately 50 MPa. The engine was operated under a constant load of 2.0 kW at a constant speed of 3000 rpm throughout the experiment. A part of the

exhaust gas was introduced into the DPF through a bypass line maintained at a constant temperature of approximately 200 °C. The superficial velocity of the exhaust gas traveling through the DPF wall was fixed at approximately 12 cm/s. The engine was stopped after some diesel particulates were trapped in the DPF, where the trapped particulate mass is linearly proportional to the engine operation time.

<figure 2 here>

Next, the DPF was regenerated at the temperature of 600 °C using a high-temperature working gas that was a mixture of nitrogen (93%) and oxygen (7%) and heated by an electric heater. During regeneration, the concentrations of CO and CO<sub>2</sub> emitted from the oxidation reaction of the trapped particulates were measured by an infrared gas analyzer.

## MICROSCOPIC VISUALIZATION SETUP

Figure 2 shows a schematic diagram of the microscopic visualization setup. A wall-flow type DPF made of SiC was cut into a small-sized DPF with a cross-sectional area of 5 × 5 mm<sup>2</sup> and a length of 7 mm, in which there is a 3 × 3 channel matrix. The alternate channels were blocked with cement at each end. After the top horizontal wall was removed, the end surfaces of the remaining vertical walls of the channels were polished using sandpaper including 10 μm silicon carbide particles. The open flow channels were covered with a 0.5-mm-thick quartz glass plate. The small DPF with the glass was installed in an insulated flow duct. Through the window, the trapping and regeneration processes were observed from both the surface and the cross-sectional views using an all-in-focus microscope (Photron Focuscope FV-100C) that provides a focal range up to 100 μm and a real-time movie at

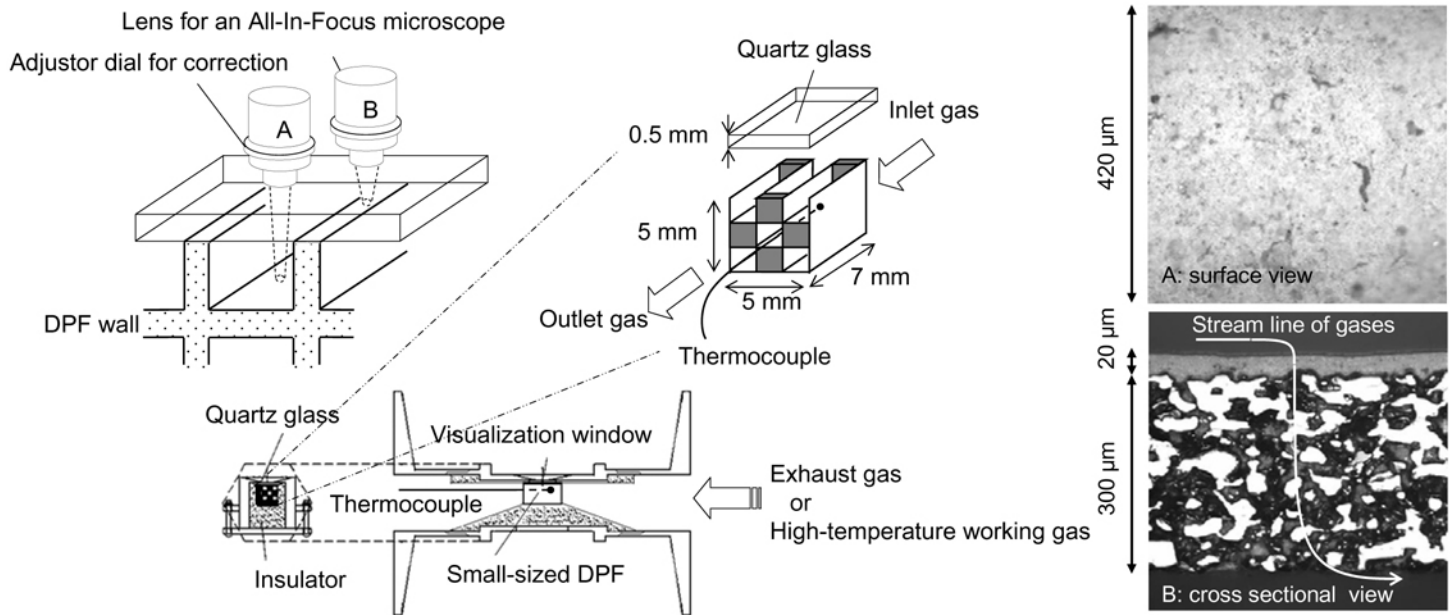


**Figure 2.** Setup for microscopic visualization of diesel particulate trapping and oxidation inside the DPF wall.

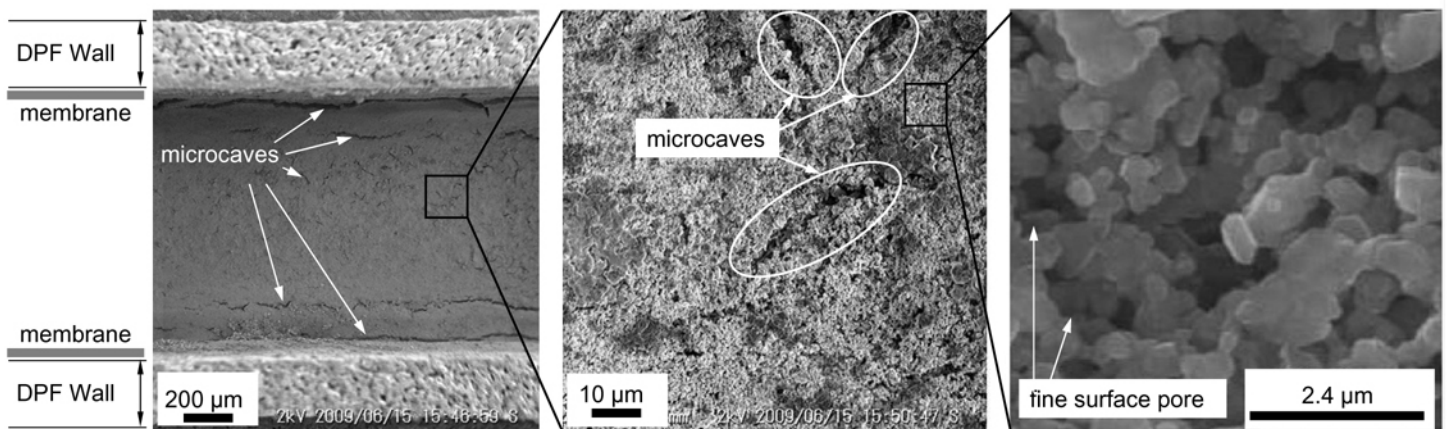


**Figure 3.** SEM images of DPMF sintered onto the conventional DPF wall (left), microcaves on the DPMF surface (middle), and fine surface pores on the DPMF surface (right).

a speed of 30 frames per second. The DPF temperature was monitored using a K-type sheathed thermocouple in the out-flow channel.

## MICROSTRUCTURE OF SIC-DPMF

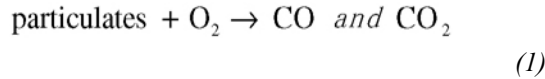
Photographs A and B on the right-hand side in Fig.2 show the surface view and the cross-sectional view of the diesel particulate catalyst-membrane filter (DPMF), respectively. The filter was made of SiC nanoparticles with a mixed size of 80 nm and 500 nm and sintered onto the conventional SiC-DPF. The DPMF has a thickness of 20 μm and a porosity of 60%, while the conventional DPF has 300 μm thickness and 42% porosity. The cross-sectional view of the DPMF on the

conventional DPF in Fig.2 is very similar to that of the soot cake trapped on it in Fig.1. In contrast, there are no nanoparticles in the surface pores. Figure 3 shows SEM images of the DPMF sintered on the in-flow channel (left), microcaves (middle), and the fine surface pores (right) on the surface of the DPMF. Because the nanoparticles were homogeneously distributed, the mean pore size of the DPMF (0.5 μm) was much smaller than that of the conventional filter (11 μm). However, some microcaves can be seen on the DPMF surface, as well as fine surface pores with a mean size of about 1 μm.

<figure 3 here>

## EVALUATION OF REACTION ACTIVITY

In order to evaluate the reaction activity of the nanoparticle membrane, a comparison between activation energies for reactions of particulates trapped on the membrane and on the conventional DPF was made, where the activation energy was obtained from the production rates of CO and CO<sub>2</sub> and the DPF temperature. Here, the particulates were assumed to be reacted with oxygen and resulted in products of CO and CO<sub>2</sub> as follows:



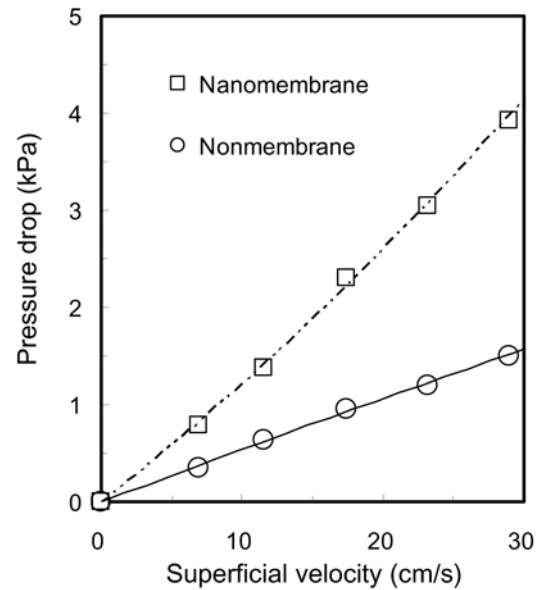
The production rate of CO and CO<sub>2</sub> can be expressed as follows:

$$\frac{d[Y_{\text{CO+CO}_2}]}{dt} = -k[Y_{\text{CO+CO}_2,\text{max}} - Y_{\text{CO+CO}_2}]^n [\text{O}_2]^m \quad (2)$$

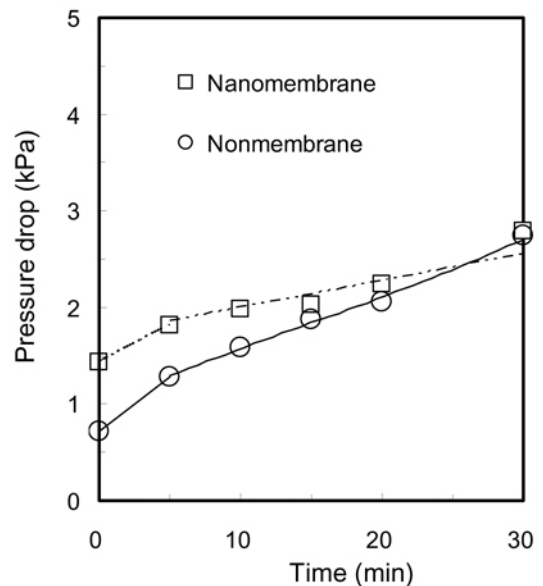
Here,  $[Y_{\text{CO+CO}_2}]$  is the summation of number of moles of CO and CO<sub>2</sub> at each time step during regeneration, while  $[Y_{\text{CO+CO}_2,\text{max}}]$  is the summation of total moles of CO and CO<sub>2</sub> from the beginning to the end of the reaction for all trapped particulates. The reaction coefficient  $k$  can be described by the Arrhenius-type reaction rate expression. As a result, the activation energy is obtained as the gradient of the following equation:

$$\ln \left[ \frac{-1}{[Y_{\text{CO+CO}_2,\text{max}} - Y_{\text{CO+CO}_2}]^n} \frac{d[Y_{\text{CO+CO}_2}]}{dt} \right] = -\frac{E}{RT} + (\ln A + m \ln [\text{O}_2]) \quad (3)$$

Here,  $E$  is the activation energy,  $A$  is the frequency factor,  $R$  is the gas constant,  $T$  is the absolute temperature of the DPF, and the last term on the right-hand side in Eq.(3) is assumed to be constant. Aleksey Yezerets *et al.* [9] reported that the above reaction was independent of any value of reaction order,  $n$ , in the range between 2/3 and 1. As a result, in the present study, the reaction order  $n$  was assumed to be unity for simplicity.



(a)



(b)

Figure 4. Pressure drops with superficial (filtration) velocity (a) and with elapsed time from the beginning of trapping (b).

## RESULTS AND DISCUSSION

### PRESSURE DROP BY MEMBRANE FILTER

Figure 4 (a) shows pressure drops for the DPMF and the conventional DPF, in the case of no soot loading. Both pressure drops increase in linear proportion to the superficial