The Effects of Filter Porosity and Flow Conditions on Soot Deposition/Oxidation and Pressure Drop in Particulate Filters

Seungmok Choi^{a*}, Kwang-Chul Oh^b, and Chun-Bum Lee^b

^aTransporation Technology R&D Center, Argonne National Laboratory

9700 South Cass Avenue, Argonne, Illinois 60439, U.S.A.

^bDiesel/hybrid Research Center, Korea Automotive Technology Institute

74, Yongjung-ri, Pungse-myun, Dongnam-gu, Chonan-si, Chungnam, 330-912, Korea

*Corresponding author: Tel.: +1-630-252-1815; Fax: +1-630-252-3443;

E-mail: schoi@anl.gov

ABSTRACT

Particulate filters are widely used in the automotive industry to reduce particulate matter (PM) produced by internal combustion engines. Wall-flow particulate filters trap PM while exhaust gas passes through the porous walls of the filter, with the pore microstructure of the porous walls affecting soot deposition, oxidation, and pressure drops during filtration and regeneration. In this study, soot deposition/oxidation behaviors were visualized in relation to the pressure drop, and the pressure drop characteristics of two particulate filters having different porosity were compared based on the results of the visualizations. It was found that the oxidation rate of the soot cake on channel walls is slower than that of the soot in the pores, due to limited oxidizer diffusion into the soot cake, which causes three-stage regeneration under the controlled regeneration regime. The high-porosity

filter offered a lower pressure drop at the same amount of soot loading, faster pressure drop recovery, and higher regeneration efficiency during controlled regeneration.

Keywords: Particulate filter, Pressure drop, Filtration, Controlled regeneration, Filter porosity

1. INTRODUCTION

The current Euro-5 emission standards for light-duty vehicles require an 80% reduction in particulate matter (PM) mass emissions relative to 2005's Euro-4 standards (i.e., to 0.005 g/km). Moreover, Euro-6 will regulate particulates larger than 23 nm in diameter to a limit of 6.0×10^{11} particles/km [1]. These standards are very difficult to be met by only optimizing in-cylinder combustion. Diesel particulate filter (DPF) systems are well known as efficient tools for controlling tail-pipe PM emissions. A DPF traps PM while exhaust gas flows through its porous ceramic walls. However, because soot loading increases the back pressure in the exhaust system, leading to low fuel economy [2], the soot needs to be removed by regeneration either periodically or continuously.

Soot deposition in a wall-flow-type DPF takes place in two distinct stages. In the first stage, the micropores in the walls are filled by particulates (depth filtration stage), which results in a rapid increase in pressure drop and evolution of filtration efficiency as a result of changes in the wall microstructure. In the second stage, a soot cake layer builds up on the incoming channel surface (the soot cake formation stage), resulting in a moderate increase in pressure drop and almost 100% filtration efficiency [3]. Because the increase in pressure drop during soot deposition is affected by such soot cake properties as packing density and Darcy permeability, which are a function of flow

conditions (i.e., flow rate, temperature, pressure), there have been attempts to characterize these soot cake properties in terms of the flow conditions. The Pe asymptotic method correlates the dimensionless Péclet number, Pe , (which is obtained by correlations among the wall velocity, the soot primary particle diameter, and the soot aggregate diffusivity) with soot cake properties [4]. Some researchers have defined “flow resistance” as a product of packing density and the Darcy permeability of soot cake, $(\rho \times k)_{\text{soot}}$, and have found that this value is correlated with temperature [5], mean free path of soot [6], and wall velocity [7]. Other research has shown that the packing density of soot cake varies between 25 and 100 kg/m³, depending on flow velocity, pressure drop, and soot loading [8]. Filter geometry (such as filter volume, cell density, and wall thickness) and pore microstructure (including porosity, pore size/size distribution, and pore connectivity) also affect soot deposition. Typically in wall-flow-type filters, a larger geometric surface area and a thinner wall will improve pressure drop. Filters have been developed having high cell density (~300 cpsi) with asymmetric cell technology and thin wall thickness (~8 mil), thanks to improvements in material strength [9-11]. It has been also been reported that hexagonal cell geometry reduces pressure drop and enhances regeneration efficiency due to an increase in filtration area [12, 13]. However, few researchers have understood the relationship between pore microstructure and filtration pressure drop. Investigators at Corning, Inc. claimed in a patent that pore microstructure specifications with high porosity (>53%), narrow pore size distribution $[(d_{50}-d_{10})/d_{50} < 0.6]$, and small mean pore diameter ($d_{50} < 20 \mu\text{m}$) are required for a better pressure drop during soot deposition [14].

Regeneration is usually classified into two ways, depending on the regeneration method (active/passive regeneration) and soot oxidation behavior (controlled/uncontrolled regeneration). Active regeneration involves controlling engine parameters or external heating hardware (such as an electric heater, burner, or microwave), while passive regeneration usually employs catalytic means (i.e., catalytic filter coatings, fuel-borne catalyst, and reactive species $[\text{NO}_2]$ generation) [15]. Controlled regeneration usually means slow soot oxidation at a low temperature (around 400°C) without a noticeable increase in the temperature in the filter. In contrast, uncontrolled regeneration employs rapid oxidation of soot at high inlet temperature (above 650°C) under high soot-loading conditions, with a sharp increase in the temperature as a result of a rapid exothermic reaction or the luminous burning flame of soot [16]. Although uncontrolled regeneration can remove soot quickly, the soot burning rate becomes uncontrolled, and the filter can be damaged because of the local high temperature, especially at transient engine operation with a sudden decrease in gas flow rate (e.g., drop-to-idle). Controlled DPF regeneration can prevent the filter from thermal failure, but the regeneration efficiency needs to be improved to minimize engine fuel consumption. Our group has reported that controlled regeneration typically involves three distinct stages in pressure drop, with a major decrease in pressure drop occurring in the second stage with the removal of soot in effective pores [17]. However, the effects of the porosity on controlled regeneration are not yet clearly understood.

In this study, we visualized soot deposition and oxidation during filtration and regeneration. On the basis of our findings, we evaluated the filtration and regeneration performance of two filters having

different pore microstructures in terms of the pressure drops across the filters. For filtration, two different flow rates represented by different Pe numbers were selected at the same inlet gas temperature and soot mass concentration, to examine the effects of soot cake properties on the increases in pressure drop for different pore microstructures. Soot loading in the filters, at four different conditions (i.e., two filter samples and two flow rates), were oxidized under controlled regeneration under the same exhaust gas flow condition (600 ppm of NO_2 and 400°C), to find the differences in regeneration processes between the soot loading conditions.

2. EXPERIMENTAL SETUP AND METHODOLOGY

2.1 Experimental Setup

A bench-scale particulate filter test system was fabricated to precisely control flow conditions, such as flow rate and temperature, during filtration and regeneration tests during engine operation. Figure 1 shows a schematic of the entire experimental setup. A 2.7 L heavy-duty, single-cylinder diesel engine was used, with exhaust gas being sampled from the main exhaust pipe of the engine. The mass flow rate passing through the particulate filter was kept constant during filtration and regeneration by controlling an automated control valve in communication with a venturi flow meter that had a proportional-integral-derivative (PID) controller. Electric heater units having a total power output of 6 kW were installed upstream of the filter to heat the inlet gas for regeneration. NO_2 gas was added downstream of the heater units to conduct a controlled regeneration at a relatively low regeneration temperature and high NO_2 concentration without using an oxidation catalyst. Soot mass

concentrations were also measured upstream and downstream of the filter by using the tapered element oscillating microbalance (TEOM) through a two-stage dilution system; the soot mass fed to the filter during filtration was calculated, based on the measured soot mass concentration and the mass flow rate of exhaust gas through the filter. The pressure drop across the filter and the temperatures at the filter inlet and outlet were measured during the entire filtration and regeneration processes.

Figure 1

Two different filter reactors serving different experimental purposes were used during the tests. One was an optically accessible filter reactor equipped with a micro-imaging system that included a stereo microscope, a high-resolution digital CCD camera, and a high-performance computer with imaging software, which enabled us to record high-magnification video images of the filter channel for filtration and regeneration (see left-hand photo in Figure 2). A bisected $\Phi 2\text{-in.} \times L6\text{-in.}$ cylindrical bare cordierite filter was installed in this optical reactor, which had a tightly sealed $3.5\text{-in.} \times 1.75\text{-in.}$ quartz window that permitted soot deposition and oxidation to be visualized on the surface of the incoming flow channel wall. The other reactor enclosed a squared $1.7\text{-in.} \times L6\text{-in.}$ filter without an optical window (shown in right-hand photo of Figure 2). This reactor was used to compare filter performance during filtration and regeneration for filters having different pore microstructures.

Figure 2

2.2 Filter Specifications

Three different wall-flow-type, non-catalyzed cordierite particulate filters were tested in this study, one for optical examination of soot deposition and oxidation and the other two to compare the filtration and regeneration performances of filters having different pore microstructures. The specifications for the filter that was placed in the optical reactor are listed in Table 1.

Table 1

Table 2 shows the specifications of the filters used for the comparisons of filtration and regeneration performance. Two filters—DPF A and DPF B—were selected to differ significantly in pore microstructure while being identical in size and geometry. The pore microstructural characteristics measured by a mercury porosimetry listed in Table 2. Mean pore size (d_{50}) is defined as the pore size when the mercury intruded 50% of total pore volume, and d_{10} and d_{90} are pore sizes when 10% and 90% of total pore volume were left during mercury intrusion, respectively. Pore size distribution was defined as $((d_{50}-d_{10})/d_{50})$. The pore size distributions of the two filters are shown in Figure 3. DPF B had a higher porosity, larger mean pore size (d_{50}), and a more uniform pore size distribution than DPF A.

Table 2

Figure 3

Clean-filter pressure drops were measured for the two filters under various flow rates in a temperature range from room temperature to 500°C; the results are plotted in Figure 4. The clean-filter pressure drop of DPF A was measured to be almost twice that of DPF B over the entire same space velocity range, because of lower gas permeability through the porous wall of DPF A caused by

the relatively low porosity and small pore size.

Figure 4

2.3 Experimental Conditions

Two different conditions for filtration were selected to take into account the flow conditions of a full-sized DPF for a light-duty diesel engine (typically $\Phi 5.66$ in. $\times$ L6 in.) and to make different soot loading conditions in terms of soot cake properties (such as packing density and permeability). The flow rates through the filters were controlled by using the filter test system, while the engine operating condition was maintained at 1,600 rpm–50% throughout the test to keep soot concentration and emissions constant for all cases. The flow conditions are listed in Table 3.

Table 3

The low and high flow-rate conditions are equivalent to the speed-load conditions of (1) 1,250 rpm and BMEP (brake mean effective pressure) of 2–3 bar and (2) 2,000 rpm and BMEP of 5–6 bar, respectively, for a typical 2.0-L turbocharged diesel engine having a typical full-sized DPF ($\Phi 5.66$ in. $\times$ L6 in.).

The “ Pe number asymptotic method” has been introduced in DPF research [4] to explain the relationship between flow condition and soot deposition properties. Pe is a non-dimensional parameter that expresses the movement of aerosols or small particles along the flow; it is defined as the ratio of convection and diffusion, as shown in Eq. (1).

$$Pe = \frac{u \cdot d_{primary}}{D_p} \quad (1)$$

Primary soot particle size ($d_{primary}$) is known to be between 25 and 40 nm in typical diesel engines, and the value of 30 nm is provided in this study. Soot aggregate-based diffusion coefficient (D_p) describes the diffusion motion soot aggregate as in Eq. (2).

$$D_p = \frac{\kappa_B \cdot T}{3\pi \cdot \mu \cdot d_{aggregate}} \cdot SCF \quad (2)$$

Soot aggregate size ($d_{aggregate}$) generally has a wide distribution and strongly depends on engine types as well as operating conditions; however, the mean diameter of 100 nm is used as a convenience reference in this study. SCF is the Stokes-Cunningham factor, which accounts for the deviation of aggregate diffusion from continuum behavior (Eq. (3)). The Knudsen number is defined in Eq. (4) for soot aggregate, based on the gas mean free path (λ) calculated by Eq. (5).

$$SCF = 1 + kn(1.257 + 0.4e^{-1.1/kn}) \quad (3)$$

$$kn = \frac{2\lambda}{d_{aggregate}} \quad (4)$$

$$\lambda = \nu \sqrt{\frac{\pi MW}{2RT}} \quad (5)$$

If Pe is smaller than unity, soot particle motion is dominated by diffusion, and the soot cake packing density decreases and soot cake permeability increases rapidly as Pe drops closer to zero. In the convection-dominated region (Pe higher than unity), the packing density and permeability of the soot cake rarely change with an increase in Pe and asymptotically become certain constant levels. To predict soot cake properties in the two flow conditions based on the Pe number asymptotic method, the wall flow velocity (U_{wall}) and average through-pore flow velocity (U_{pore}) were calculated, and by

using U_{wall} and U_{pore} as inputs in Eq. (1) in the velocity term u , the Pe number of wall flow (Pe_{wall}) and the Pe number of pore flow (Pe_{pore}) were finally calculated, as listed in Table 4.

Table 4

U_{wall} is defined as the division of volumetric flow rate through the filter by filter geometric surface area (GSA); accordingly, Pe_{wall} is expected to have a closer relationship with the soot cake properties formed on the wall surface. Similarly, U_{pore} is defined as the division of volumetric flow rate through the filter by filter geometric pore area (defined as the GSA multiplied by porosity); consequently, the calculated Pe_{pore} is suitable for explaining the soot properties in pores. Pe_{wall} values are smaller than unity (diffusion-dominated) for all cases, and the Pe_{wall} of low-flow conditions is much lower than that of high-flow conditions. In other words, the soot cake formed on the wall under low-flow conditions will have lower packing density and higher permeability than the soot cake formed under high-flow conditions. The Pe_{pore} values of DPF A are all higher than unity (convection-dominated), which indicates the soot deposited in pores has similar packing density and permeability for the two flow conditions. In DPF B, however, the soot deposited in pores will have different properties because the low-flow condition is diffusion-dominated ($Pe_{pore} < 1$) and the high-flow condition is convection-dominated ($Pe_{pore} > 1$). After soot loading in four different cases, regeneration tests were conducted to examine the effects of soot-deposition characteristics and filter specifications on regeneration performance under the same conditions ($NO_2 = 600$ ppm, temperature = 400~440°C, and mass flow rate = 17 kg/h).

2.4 Experimental Methodology

To compare the pressure drop level caused only by soot loading (ΔP_{soot}), ΔP_{soot} was obtained by subtracting the clean-filter pressure drop (ΔP_{clean}) from the measured total pressure drop across the filter (ΔP_{total}). ΔP_{clean} and ΔP_{soot} can be expressed by a 0-D pressure drop equation for wall-flow-type filters under the assumptions of uniform flow and thermal conditions, as shown in Eq. (6) and Eq. (7), respectively [18].

$$\Delta P_{clean} = \frac{\mu Q (\alpha + w_s)^2}{2V_{trap}} \left[\frac{w_s}{k_0 \alpha} + \frac{8FL^2}{3\alpha^4} \right] + \frac{\rho Q^2 (\alpha + w_s)^4}{V_{trap}^2 \alpha^2} \left[\frac{\beta w_s}{4} + 2\zeta \left(\frac{L}{\alpha} \right)^2 \right] \quad (6)$$

$$\Delta P_{soot} = \frac{\mu Q (\alpha + w_s)^2}{2V_{trap}} \left[\frac{w_s}{k_{soot, wall} \alpha} + \frac{1}{2k_{soot, cake}} \ln \left(\frac{\alpha}{\alpha - 2w} \right) + \frac{4FL^2}{3} \left(\frac{1}{(\alpha - 2w)^4} - \frac{1}{\alpha^4} \right) \right] \quad (7)$$

ΔP_{clean} depends on flow conditions, filter geometry, and porous wall properties. ΔP_{soot} is a function of soot cake properties, as well as flow conditions and filter geometry. In the ΔP_{clean} equation (Eq. (6)), volume flow rate (Q), viscosity (μ), and density (ρ) are the parameters that depend on flow conditions, and, by dividing both the left- and right-hand sides of Eq. (6) by $\mu Q/V_{trap}$, we can obtain a normalized clean-filter pressure drop that has a linear correlation with $\rho Q/\mu$, as shown in Eq. (8):

$$\frac{\Delta P_{clean} V_{trap}}{\mu Q} = \frac{(\alpha + w_s)^2}{2} \left[\frac{w_s}{k_0 \alpha} + \frac{8FL^2}{3\alpha^4} \right] + \frac{\rho Q}{\mu} \frac{(\alpha + w_s)^4}{V_{trap} \alpha^2} \left[\frac{\beta w_s}{4} + 2\zeta \left(\frac{L}{\alpha} \right)^2 \right] \quad (8)$$

The linear coefficients A and B are intrinsic values, depending on filter geometry and porous wall properties, which are not affected by flow conditions. Coefficient A includes the Darcy permeability of the DPF wall and channel friction, and coefficient B stands for the Forchheimer term of the porous wall and contraction and expansion losses. The clean-filter pressure drop coefficients A and B can be

obtained from linear fitting of $\rho Q/\mu$ and normalized ΔP_{clean} , which are obtained from clean-filter pressure drop tests conducted under a wide range of temperature and flow rate conditions. Figure 5 shows the linear fitting of the measured ΔP_{clean} data and the clean-filter pressure drop coefficients of the filter used in this study. Once the coefficients A and B of a filter are obtained, one can easily calculate ΔP_{clean} only from the measured flow conditions during filtration and regeneration, and ΔP_{soot} can be separated by subtracting ΔP_{clean} from the measured ΔP_{total} .

Figure 5

ΔP_{soot} is also normalized in the same way, by dividing both sides of Eq. (7) by $\mu Q/V_{trap}$, resulting in Eq. (9):

$$\frac{\Delta P_{soot} V_{trap}}{\mu Q} = (\alpha + w_s)^2 \left[\frac{w_s}{k_{soot,wall} \alpha} + \frac{1}{2k_{soot,cake}} \ln \left(\frac{\alpha}{\alpha - 2w} \right) + \frac{4FL^2}{3} \left(\frac{1}{(\alpha - 2w)^4} - \frac{1}{\alpha^4} \right) \right] \quad (9)$$

On the right-hand side of Eq. (9), parameters related only to soot cake properties and filter geometries remain. This means that the normalized ΔP_{soot} is independent of the flow conditions, as long as the soot cake parameters are not dependent on the flow conditions. Direct comparison of the pressure-drop level due to soot loading at different flow conditions is possible with normalized ΔP_{soot} , and it is also beneficial to evaluate the relationship between the visualized soot deposition/oxidation and the pressure drop.

Figure 6 shows the traces of measured ΔP_{total} , normalized ΔP_{soot} , and filter inlet temperature during filtration, heat-up, and regeneration of DPF B under the high-flow condition. After about 0.6 g/L of soot loading, the exhaust gas is heated to 400°C by using an electric heater, and then for regeneration, NO₂ gas is supplied after the filter is fully heated up until the NO₂ concentration at the filter inlet is

600 ppm. The normalized ΔP_{soot} dropped rapidly at initial heat-up, which is likely the result of the separation of volatile fractions from the trapped soot, and it soon became flat because there is no soot oxidation before the addition of NO₂ gas. As can be seen from the trace of measured ΔP_{total} , the increase in pressure drop as a result of soot loading during filtration and regeneration is difficult to estimate and compare between different filters and different flow conditions by the measured ΔP_{total} because it fluctuates with changes in flow condition and includes ΔP_{clean} and ΔP_{soot} in it. In the trace of normalized ΔP_{soot} , the fluctuations shown in the measured ΔP_{total} are not found because the normalized ΔP_{soot} is independent of the flow conditions and only depends on the soot cake properties. And because this value indicates 0 when the filter is clean, the increase in pressure drop caused by soot cake can be evaluated more precisely during filtration and regeneration.

Figure 6

Regeneration efficiency can be defined as the ratio of normalized ΔP_{soot} at a certain time after the start of regeneration and the initial normalized ΔP_{soot} , as shown in Eq. (10):

$$\text{Regen. efficiency (\%)} = \frac{\text{Norm. } \Delta P_{soot} \text{ at certain time}}{\text{Initial Norm. } \Delta P_{soot}} \times 100 \quad (10)$$

3. SOOT DEPOSITION AND OXIDATION BEHAVIOR UNDER CONTROLLED REGENERATION

The behavior of soot deposition in the filter channel during filtration was visualized at the micro-scale, as was the pressure drop across the filter. Three distinct consecutive filtration stages were found in the normalized ΔP_{soot} : depth filtration, transitional filtration, and soot cake formation (all of

which were well known from previous research). The left column of the Figure 7 shows the normalized ΔP_{soot} trace as a function of soot loading mass in g/L and the corresponding soot deposition images (visualized from the top of the channel surface), as well as schematic representations that show cross sections of the filter wall at certain filtration times (marked on the pressure drop trace) shown on the graph.

During depth filtration, soot particles start to fill the effective pores first, with a steep increase in pressure drop. Many black spots are shown in image ② (visualized at the end of depth filtration), which indicate that the effective pores are almost fully filled with soot. Transition filtration is initiated after a majority of the effective pores are clogged with soot, at which point the pressure drop gradually increases at a lower rate until soot cake filtration begins. During transition filtration, more soot particles are deposited around the clogged pores (black spots in image ②) and spread out to the rest of the channel wall. It is likely that soot particles in the flow are initially attached to the soot deposited in the pores and eventually form soot-hills on the top. Soot cake formation (image ③) begins before the channel surface is fully covered by soot cake, and during that process, a moderate and linear increase in pressure drop is observed. A soot-loading rate of about 1.8 g/L results in a soot cake layer that fully covers the surface of the channel wall, as shown in image ④, under given flow conditions and filter specifications.

Detailed soot oxidation behaviors were observed during the controlled regeneration. The filter was fully heated to the target temperature before the start of regeneration to avoid differences in soot oxidation caused by local differences in temperature and the effects of heat transfer. The right-hand

column of Figure 7 shows the normalized ΔP_{soot} trace plotted against the elapsed time from the start of regeneration and the corresponding soot oxidation images (visualized from the top of the channel surface) and schematic representations (cross sections of the filter wall) at several controlled regeneration times (marked on the pressure drop trace) shown on the graph. Three distinct regeneration stages were identified, each of which shows a linear decrease in normalized ΔP_{soot} .

In the first regeneration stage (①→②), there is a noticeable decrease in pressure drop; however, no change in the soot cake is observed from the image visualized at the top of the channel surface (see the images ① and ②). It is likely that soot oxidation in the first stage removes mainly the soot particles deposited in the pores and decreases the soot cake thickness on the wall to a certain degree, while the wall stays covered by soot cake and the pores remain closed by soot.

Further soot oxidation finally starts to open the effective pores by removing the soot from them, and the pressure drop decreases rapidly because wall permeability increases quickly. In this second stage of regeneration, a continuous opening of pores leads to a steep and linear decrease in the pressure drop (②→③→④). Several small holes in the soot cake shown by the white spots in image ③ (taken at the middle of the second stage) indicate open effective pores; at the end of the stage, almost all of the effective pores are believed to be opened (image ④), and the decrease in pressure drop slows down. After the second stage, soot cake remains only on the wall surface and continues to oxidize until the filter becomes clean. In this third stage (④→⑤→⑥), the pressure drop decreases gradually, with the lowest gradient among the three stages. The presence of normalized ΔP_{soot} in this period is only due to the soot cake remaining on the channel surface,

because the micro-pores have been cleaned at the second stage. Note that the thickness of the soot cake on the channel surface continuously decreases by oxidation throughout the entire regeneration process.

Figure 7

Our observations show that (1) the oxidation rate of the soot in pores is higher than that of the soot cake on the channel surface and (2) the absolute amount of soot deposited in the pores is smaller than that of soot cake. Thus, soot cake remains on the channel surface after complete removal of soot from the pores during the second stage. The difference in soot oxidation rates in pores and on the surface can be explained by the difference in oxidizer transport. For the soot in pores where forced convection through the pores can easily transfer oxidizer, the soot oxidation rate is limited by the chemical reaction rate. But for the majority of soot cake deposited on the channel surface, where the oxidation should be dominated by surface diffusion from the channel gas flow, soot oxidation is limited by oxidizer transport via Knudsen diffusion [19]. This mechanism is expected to be effective for non-catalyzed substrates with the controlled regeneration condition. For catalyzed filters, the soot oxidation behavior can be different because of the “back diffusion” [16] of the higher concentration of oxidizer through the wall surface to the bottom of soot cake. The removal of soot cake remaining at the third regeneration stage takes longer and requires more energy input to the filter, even though the contribution of soot cake to the pressure drop is rather small. Therefore, the regeneration strategy could be optimized to increase energy efficiency by controlling the total operating time of regeneration near the end of the second stage.

4. COMPARISON OF PRESSURE DROP CHARACTERISTICS DURING FILTRATION AND REGENERATION

4.1 Pressure Drop Characteristics during Filtration: Effects of Porosity and Flow Conditions

In this section, we compare the characteristics of pressure drop build-up during the filtration of two particulate filters having different porosities at two different flow rates. The traces of normalized ΔP_{soot} for four different cases are plotted as a function of soot loading mass in Figure 8. The transition timing from depth filtration to soot cake formation is defined as the intersection of linear fitting lines of two filtration stages in a normalized ΔP_{soot} trace. On the basis of this criterion, the amounts of soot loading mass during depth filtration and soot cake formation were measured, and normalized ΔP_{soot} levels at the end of depth filtration were obtained. The slope of the linear fitting line represents the level of pressure drop build-up per unit mass of soot loading, and it is defined as the “pressure drop potential” in this study. The pressure drop potentials were obtained for depth filtration and soot cake formation. All of the filtration parameters for four different cases are listed in Table 5.

Figure 8

Table 5

During depth filtration, the pressure drop associated with DPF A (low porosity) was quite different from that associated with DPF B (high porosity). DPF B, a high-porosity filter, can contain 3~4 times more soot in the pores, with a potential pressure drop of about one-third to one-fourth that of DPF A.

Consequently, normalized ΔP_{soot} levels at the end of depth filtration were measured to be similar between the two filters, in that the high-porosity filter has higher soot loading capacity with lower pressure-drop potential. The difference in flow rate did not significantly affect the pressure drop during depth filtration and soot loading mass of the DPF A, but in DPF B, the low-flow rate showed a higher pressure drop potential with lower in-depth soot loading mass than the high-flow rate. This behavior can be explained from the Pe_{pore} values listed in Table 4. The flow conditions of the soot particles in pores of the low-porosity filter (DPF A) were convection-dominated ($Pe_{pore} > 1$) for both low and high flow rates, and similar soot loading mass and pressure drop potential during depth filtration were measured. However, in the high-porosity filter (DPF B), the soot deposition characteristic in pores was diffusion-dominated ($Pe_{pore} < 1$) for the low-flow rate, but it became convection-dominated ($Pe_{pore} > 1$) for the high-flow rate. Under the low-flow condition, soot loading mass was lower and the pressure drop potential was higher than for the high-flow condition. Therefore, the convection-dominated flow condition during depth filtration can offer better soot deposition characteristics—a lower pressure drop with higher in-depth soot loading for the high-porosity filter (DPF B).

During soot cake formation, when the soot cake starts to build up on the wall surface, the increase in the pressure drop of DPF A under the high-flow condition was exceptionally high among the four cases. The pressure drop potentials of the other three cases stayed in the less than 20% range. This result is not well correlated with Pe_{wall} of the four cases, which indicates that the average flow conditions of the soot particles are all diffusion-dominated ($Pe_{wall} < 1$), and so soot particles are

expected to be more diffusive in low-flow conditions (lower Pe_{wall}). Thus, Pe_{wall} calculated from the average wall flow velocity (U_{wall}) cannot represent the pressure drop characteristics during soot cake formation for filters with different porosity. The pressure drop will depend on local soot cake properties and the flow condition. Simple classification of soot cake into two regions, as shown in Figure 9, can provide better explanations for the effects of local differences on pressure drop. Soot in effective flow regions (illustrated in gray) is defined as in-depth soot and soot cake formed on top of the effective pores, where flows are actually passing through. The local soot cake properties and flow velocities at this region will dominate the pressure drop during filtration.

Figure 9

Among the four cases, DPF A with the high flow rate case allowed the highest local-flow velocity passing through the effective pores, as was reflected to the highest value of Pe_{pore} (3.219), while Pe_{pore} values for the other cases were lower and close to unity. The local Pe number at the entrances of the effective pores should be between Pe_{wall} and Pe_{pore} . Thus, the local Pe values at the entrance of the effective pores are likely to become greater than unity for DPF A with the high flow rate case, which, in turn, forms soot cake with high packing density and low permeability under the convection-dominated flow condition.

The results showed that the high-porosity filter offers a lower pressure drop increase during filtration. And the pressure drop increase due to soot trapping can be minimized by controlling the flow condition through the filter wall to have Pe_{pore} greater than unity (convection-dominated in-depth flow) and, at the same time, close to unity (diffusion-dominated flow) at the entrance of the

effective pore. The filter geometry (i.e., cell density, wall thickness, filter diameter, and length) can be determined to meet the Pe_{pore} criteria.

4.2 Pressure Drop Characteristics during Controlled Regeneration: Effects of Porosity and Soot Loading Condition

Controlled regeneration experiments were conducted by using the four different filtration cases (explained in section 4.1) at identical flow conditions (mass flow rate = 17 kg/h, NO_2 = 600 ppm, and inlet temperature = 400~440°C; see Table 3). The changes in normalized ΔP_{soot} for the four cases were evaluated during controlled regeneration based on the soot oxidation behavior found in the visualization study (section 3). The controlled regeneration of the four different filter samples clearly showed three regeneration stages, as indicated by the traces of normalized ΔP_{soot} in Figure 10. As explained in section 3, the pressure drop decreased mainly during the second regeneration stage with continuous opening of the effective pores. After the second stage, soot remained only on the wall surface and contributed little to the pressure drop. Therefore, regeneration quality until the end of the second regeneration stage is important. To evaluate the regeneration quality until the end of the second regeneration stage, the time required until the end of the second stage, the normalized ΔP_{soot} drop rate during the second stage, and the normalized ΔP_{soot} level and regeneration efficiency (Eq. (10)) at the end of the second stage were calculated for the four cases listed in Table 6.

Figure 10

Table 6

The results showed that the time required to finish the second regeneration stage was similar for all of the cases (about 900~1,100 s). This finding means that the time required to completely oxidize (remove) soot in the effective flow regions (Figure 9) is similar under the same regeneration flow conditions (i.e., flow rate, temperature, and oxidizer type and concentration), and the effects of the filter microstructure, total soot loading mass, and the soot cake properties (i.e., packing density, permeability) are minor. Especially with the DPF A in the low flow rate case, it took about the same time to completely oxidize (remove) soot in the effective flow regions, even with about half of the total soot loading mass of the other cases. The implication is that the actual soot mass oxidation (removal) rate during the second regeneration stage becomes faster in proportion with the soot mass in the effective flow regions. Again, the actual soot mass oxidation (removal) rate is also proportional to the total amount of soot loaded in the filter, because the amount of soot in the effective flow regions increases as more total soot is loaded in the filter. This soot oxidation behavior is due to the chemically limited soot oxidation rate in controlled regeneration with the forced convection flow of sufficient oxidizer through the soot in the effective flow regions, as explained in section 3. The slowest normalized ΔP_{soot} drop of DPF A with the low flow rate case can be explained in the same manner. As compared to DPF A in a high flow rate case, the total soot loading mass of the low flow rate case is about half, which results in a mass oxidation rate of the soot in the effective flow region of about half. The normalized ΔP_{soot} drop rates of the other three cases were in similar ranges because the total soot loading masses of those cases are similar to those shown in Table 6.

The normalized ΔP_{soot} at the end of the second regeneration stage is due to the soot cake remaining on the wall, and this value is important because it takes a longer time to remove the remaining soot cake and recover the pressure drop at the third stage. The results showed that higher normalized ΔP_{soot} was measured in DPF A (low porosity) than in DPF B (high porosity). Again, if we compare the DPF A low and high flow rate cases, the value was about twice as large for the high flow rate case because the total soot loading mass at the high flow rate case was twice as high as that of the low flow rate case and will therefore leave about twice the amount of soot cake remaining on the wall after the second stage. However, in terms of the regeneration efficiency at the end of the second stage, the regeneration efficiency was related only to filter porosity, which was about 70% for DPF A and about 90% in DPF B, regardless of the total soot loading mass and the differences in filtration conditions. The higher regeneration efficiency (less soot cake remaining on the wall) of the high-porosity filter can be clearly explained in the schematic drawings of soot oxidation behavior in the low- and high-porosity filters shown in Figure 11. The soot in the effective flow regions is completely removed during the first and second stages at a higher rate (chemically limited). In contrast to the soot in the effective flow regions, the soot cake on the wall is oxidized from the top surface at a lower rate, limited by Knudsen diffusion, and thinned out throughout the regeneration. In the high-porosity filter, the volume of effective flow regions is larger and the area of the top wall surface is smaller than those of the low-porosity filter. And the soot cake thickness in the high-porosity filter is thinner at the same soot loading mass because of the higher amount of in-depth soot than in the low-porosity filter. (The differences of soot cake properties are ignored here, because

those effects are minor.) As a result, much less soot cake remains in the high-porosity filter after the second-stage regeneration, as illustrated in the Figure 11, the result of which is the lower normalized ΔP_{soot} and higher regeneration efficiency of the high-porosity filter at the end of the second regeneration stage.

Figure 11

Therefore, a high-porosity filter offers higher regeneration efficiency and thus offers an advantage in terms of a faster rate of soot mass oxidation (due to the larger volume of effective flow regions) and higher regeneration efficiency (because less soot cake remains on the wall after the second regeneration stage).

5. CONCLUSIONS

In this study, the soot deposition and oxidation behaviors and the effects of these parameters on pressure drop during filtration and controlled regeneration were examined by the microscopic visualization of the incoming channel in a non-catalyzed cordierite filter. The pressure drop characteristics of two filters with different porosity were compared and analyzed during filtration and controlled regeneration. A dimensionless parameter, normalized ΔP_{soot} , was derived from a 0-D pressure drop equation, to compare the level of pressure drop caused only by soot loading from the measured total pressure drop across the filter, regardless of flow conditions. Three-staged regeneration was identified from the visualization and measured pressure drop. A significant decrease in pressure drop occurred during second-stage regeneration, with the removal of soot in the

effective flow regions and the opening of effective pores. Soot cake remained on the wall surface after the second stage with little contribution to the pressure drop. Filtration experiments with two different filters (low and high porosity) under two flow rate conditions (low and high flow rates) showed that the high-porosity filter lowers the pressure drop increase due to soot loading at the same level of soot loading, and the pressure drop increase by soot loading can be minimized by controlling the flow condition through the filter wall such that the Pe_{pore} is greater than unity (convection-dominated in-depth flow), but close to unity at the same time (diffusion-dominated flow at the entrance of the effective pore). Under controlled regeneration, a high-porosity filter is also preferred for quicker pressure drop recovery as a result of a faster soot mass oxidation rate (which is associated with a larger volume of effective flow regions) and higher regeneration efficiency (because less soot cake remained on the wall after the second regeneration stage).

The results obtained in this research would likely be valid only with the bare filter and controlled regeneration with sufficient oxidizer concentration. Further investigations are required to determine the regeneration in catalyzed filters, in which the back diffusion from the bottom of the soot cake layer occurs.

6. ACKNOWLEDGEMENTS

The submitted manuscript has been created by UChicago Argonne, LLC, Operator of Argonne National Laboratory (“Argonne”). Argonne, a U.S. Department of Energy Office of Science laboratory, is operated under Contract No. DE-AC02-06CH11357. The U.S. Government retains for

itself, and others acting on its behalf, a paid-up nonexclusive, irrevocable worldwide license in said article to reproduce, prepare derivative works, distribute copies to the public, and perform publicly and display publicly, by or on behalf of the Government. This work was supported by the U.S. Department of Energy, Office of Vehicle Technologies, and Korea Automotive Technology Institute (KATECH) under Contract No. C1000301.

7. REFERENCES

- [1] DieselNet website: www.dieselnet.com/standards/eu/ld.php, Oct. 22, 2013.
- [2] Lapuerta, M., J. Rodríguez-Fernández, and F. Oliva, “Effect of Soot Accumulation in a Diesel Particle Filter on the Combustion Process and Gaseous Emissions,” *Energy*, 47(1), 543–552, 2012.
- [3] Tandon, P., A. Heibel, J. Whitmore, N. Kekre, and K. Chithapragada, “Measurement and Prediction of Filtration Efficiency Evolution of Soot Loaded Diesel Particulate Filters,” *Chemical Engineering Science*, 65, 4751–4760, 2010.
- [4] Konstandopoulos, A.G., E. Skaperdas, and M. Masoudi, “Microstructural Properties of Soot Deposits in Diesel Particulate Traps,” *SAE paper* 2002-01-1015, 2002.
- [5] Pattas, K.N., A.M. Stamatelos, G.C. Koltsakis, I.P. Kandylas, and W. Mustel, “Computer Aided Engineering in the Design of Catalytically Assisted Trap Systems,” *SAE paper* 970472, 1997.
- [6] Versaevel P., H. Colas, C. Rigaudéeau, R. Noirot, G. Koltsakis, and A. Stamatelos, “Some Empirical Observations on Diesel Particulate Filter Modeling and Comparison between Simulations

and Experiments,” *SAE paper* 2000-01-0477, 2000.

[7] Yoon, C.S., S.H. Song, and K.M. Chun, “Measurement of Soot Mass and Pressure Drop Using a Single Channel DPF to Determine Soot Permeability and Density in the Wall Flow Filter,” *SAE paper* 2007-01-0311, 2007.

[8] Koltsakis, G.C., A. Konstantinou, O.A. Haralampous, and Z.C. Samaras, “Measurement and Intra-Layer Modeling of Soot Density and Permeability in Wall-flow Filters,” *SAE paper* 2006-01-0261, 2006.

[9] Aravelli, K., and A. Heibel, “Improved Lifetime Pressure Drop Management for Robust Cordierite (RC) Filters with Asymmetric Cell Technology (ACT),” *SAE paper* 2007-01-0920, 2007.

[10] Boger, T., J. Jamison, J. Warkins, N. Golomb, C. Warren, and A. Heibel, “Next Generation Aluminum Titanate Filter for Light Duty Diesel Applications,” *SAE paper* 2011-01-0816, 2011.

[11] Boger, T., S. He, T. Collins, A. Heibel, D. Beall, and C. Remy, “A Next Generation Cordierite Diesel Particle Filter with Significantly Reduced Pressure Drop,” *SAE paper* 2011-01-0813, 2011.

[12] Tsuneyoshi, K., and Y. Kazuhiro, “A Study on the Cell Structure and the Performances of Wall-Flow Diesel Particulate Filter,” *Energy*, 48(1), 492–499, 2012.

[13] Tsuneyoshi, K., and Y. Kazuhiro, “Experimental Study of Hexagonal and Square Diesel Particulate Filters under Controlled and Uncontrolled Catalyzed Regeneration,” *Energy*, 60, 325–332, 2013.

- [14] Merkel, G.A., and T. Tao, "Narrow Pore Size Distribution Cordierite Filters with Reduced Pressure Drop," *United States Patent Application Publication*, US2007/0107397 A1.
- [15] Konstandopoulos, A.G., M. Kostoglou, E. Skaperdas, E. Papaioannou, D. Zarvalis, and E. Kladopoulou, "Fundamental Studies of Diesel Particulate Filters: Transient Loading, Regeneration, and Aging," *SAE paper* 2000-01-1016, 2000.
- [16] Haralampous, O.A., G.C. Koltsakis, and Z.C. Samaras, "Reaction and Diffusion Phenomena in Catalyzed Diesel Particulate Filters," *SAE paper* 2004-01-0696, 2004.
- [17] Choi, S., and K. Lee, "Detailed Investigation of Soot Deposition and Oxidation Characteristics in a Diesel Particulate Filter Using Optical Visualization," *SAE paper* 2013-01-0528, 2013.
- [18] Konstandopoulos, A.G., "Flow Resistance Descriptors for Diesel Particulate Filters: Definitions, Measurements and Testing," *SAE paper* 2003-01-0846, 2003.
- [19] Stanmore, B.R., J.F. Brilhac, and P. Gilot, "The Oxidation of Soot: A Review of Experiments, Mechanisms, and Models," *Carbon*, 39, 2247–2268, 2001.
- [20] Lee, K., S. Choi, H. Seong, Y. Kameya, and H. Lee, "Particulate Emissions Control by Advanced Filtration Systems for GDI Engines," DOE Annual Merit Review & Peer Evaluation Meeting, May 15, 2013.

NOMENCLATURE

A clean filter pressure drop coefficient (y-intersect)

B	clean filter pressure drop coefficient (slope)
$d_{aggregate}$	soot aggregate diameter
d_{10}	pore sizes when 10% of total pore volume is left during mercury intrusion
d_{50}	mean pore diameter
d_{90}	pore sizes when 90% of total pore volume is left during mercury intrusion
DPF	diesel particulate filter
D_p	soot-aggregate-based diffusion coefficient
$d_{primary}$	primary soot particle diameter
F	factor (=28.454)
GSA	filter geometric surface area
k_0	clean filter wall permeability
k_B	Boltzmann's constant
$k_{soot,wall}$	permeability of soot in wall
$k_{soot,cake}$	permeability of soot cake
Kn	Knudsen number
L	filter outer diameter
MW	gas molecular weight
Pe	Péclet number
Pe_{wall}	average Péclet number calculated by U_{wall}
Pe_{pore}	average Péclet number calculated by U_{pore}

PM	particulate matter
R	universal gas constant
SFC	Stokes-Cunningham factor
T	gas temperature
TEOM	tapered element oscillating microbalance
Q	exhaust volumetric flow rate
U_{wall}	average wall velocity
U_{pore}	average flow velocity through pores
u	velocity to calculate Pe
V_{trap}	filter volume
w	soot cake thickness
w_s	filter wall thickness

Greek Letters

α	honeycomb filter cell size
β	Forchheimer coefficient
ΔP_{clean}	clean filter pressure drop
ΔP_{soot}	pressure drop by soot loading
ΔP_{total}	total pressure drop across filter
ζ	contraction/expansion loss coefficient
λ	exhaust gas-mean-free-path

μ	exhaust dynamic viscosity
ν	exhaust kinematic viscosity
ρ	exhaust gas density

FIGURES

Figure 1 Schematic of the bench-scale particulate filter test system

Figure 2 Bench-scale particulate filter reactors with (left) and without (right) an optical window

Figure 3 Pore size distributions of the two particulate filters measured by mercury intrusion porosimetry

Figure 4 Clean-filter pressure drops of the two particulate filters

Figure 5 Clean-filter pressure drop coefficients of the two particulate filters

Figure 6 Traces of measured ΔP_{total} , normalized ΔP_{soot} , and filter inlet temperature during filtration, heat-up, and

regeneration (DPF B with high-flow case)

Figure 7 Visualized soot deposition and regeneration behavior [20]

Figure 8 Traces of normalized ΔP_{soot} plotted against the soot loading mass for four different filtration cases

Figure 9 Classification of soot in a filter: soot cake on the wall (black) and soot in effective flow regions (gray)

Figure 10 Traces of normalized ΔP_{soot} during controlled regeneration for four different soot loading cases

	Low Porosity	High Porosity
First stage		
Second stage		
Third stage		

Figure 11 Schematic drawings for comparison of soot oxidation behaviors in low-porosity and high-porosity filters

TABLES

Table 1 Specifications of the particulate filter for optical examination

Description	Optical DPF
Size	$\Phi 2\text{-in.} \times \text{L6-in. half-cut}$
Cell density (cpsi)	200
Wall thickness (inch)	0.012
Porosity (effective) (%)	48
Mean pore size (d_{50}) (μm)	13

Table 2 Specifications of particulate filters for pore microstructure comparison

Description	DPF A	DPF B
Size	1.7 in. square $\times$ 6 in.	
Cell density (cpsi)	200	
Wall thickness (in.)	0.012	
Porosity (effective) (%)	20.89	47.05
Mean pore size (d_{50}) (μm)	7.242	11.948
Pore size distribution		
$((d_{50}-d_{10})/d_{50})$	0.79	0.41
d_{10} (μm)	1.51	7.03
d_{90} (μm)	21.13	25.11

Table 3 Flow conditions for filtration and regeneration

Filtration	
Soot conc. (mg/m^3 @ std.)	9
DPF inlet temp. ($^{\circ}\text{C}$)	150~200
Mass flow rate (kg/h)	7~8 (low flow) / 18~20 (high flow)
Regeneration	
NO_2 concentration (ppm)	600
DPF inlet temp. ($^{\circ}\text{C}$)	400~440
Mass flow rate (kg/h)	17

Table 4 Pe number of each condition

	U_{wall} (cm/s)	Pe_{wall}	U_{pore} (cm/s)	Pe_{pore}
DPF A, Low Flow	0.777	0.236	3.719	1.131
DPF A, High Flow	2.212	0.672	10.587	3.219
DPF B, Low Flow	0.892	0.271	1.894	0.576
DPF B, High Flow	2.233	0.679	4.745	1.442

Table 5 Comparison of filtration characteristics of four filtration cases

	DPF A Low Flow	DPF A High Flow	DPF B Low Flow	DPF B High Flow
Total soot loading (g/L)	0.313	0.631	0.540	0.598
Soot loading during depth filtration (g/L)	0.034	0.033	0.118	0.163
Soot loading during soot cake formation (g/L)	0.279	0.598	0.422	0.435
Norm. ΔP_{soot} at the end of depth filtration	1,991,997	1,801,369	2,169,048	1,770,085
ΔP potential by depth filtration (Norm. $\Delta P_{soot}/(g/L)$)	43,796,817	42,837,006	18,372,021	10,518,552
ΔP potential by soot cake (Norm. $\Delta P_{soot}/(g/L)$)	2,755,172	4,772,154	3,079,911	2,595,161

Table 6 Comparison of controlled regeneration characteristics of four filtration cases

	DPF A Low Flow	DPF A High Flow	DPF B Low Flow	DPF B High Flow
Total soot loading (g/L)	0.313	0.631	0.540	0.598
Required time to the end of 2 nd stage (second)	1,129	1,013	1,104	864
ΔP drop rate during 2 nd stage (Norm. $\Delta P_{soot}/sec.$)	-1,064	-2,141	-2,148	-2,611
Norm. ΔP_{soot} at end of 2 nd stage (regeneration efficiency)	423,182 (71.8%)	827,054 (71.5%)	294,481 (85.8%)	166,154 (91.5%)