

A Study of Active and Passive Regeneration Using Laboratory Generated Soot on a Variety of SiC Diesel Particulate Filter Formulations

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ABSTRACT

In this study an attempt to understand and demonstrate the effects of various washcoat technologies under active and passive regeneration conditions was performed. Six different formulations, on 1.0" D. × 3.0" L. SiC wall flow filters at the laboratory level were used at various test conditions, including variable NO₂/NO_x ratios and O₂ concentrations. Samples were regenerated using active and passive conditions to evaluate regeneration rates and the potential impact of regeneration at the vehicle level. Results were applied to vehicle operating conditions to determine passive functionality and potential benefits. Active regenerations at 2% O₂ and 5% O₂ showed no significant difference in time to complete regeneration and soot burn rates. Active regenerations performed at 1% O₂ and 5% O₂ concentration showed that the regeneration temperature was shifted by approximately 50°C. Active soot burn rates were mainly governed by the higher inlet temperature and showed minor differences between Pt containing washcoats and non Pt containing samples. Pt containing samples provided a much cleaner reaction in that even a low loading of Pt converted > 99% of the soot to CO₂. The Pd only washcoat also was able to achieve mostly CO₂ as a product. For both non PGM containing washcoats and blank substrate there was a 60:40 ratio of CO:CO₂. Overall, a Cu/zeolite containing sample showed similar results to the blank substrate, in that the regeneration capability was approximately equal to a blank substrate. The significant difference was observed when comparing the soot burn rates from the low and high soot loaded samples. As the soot loading was nearly doubled for each sample the soot burn rate increased by nearly the same value under active conditions only. Comparing the passive regeneration rates for the different cases, the 0.5 blend of NO

₂/NO_x was on average 2.5 faster then what was observed for the standard or lower NO₂/NO_x ratio of 0.15. Results show that if operating in an appropriate temperature range with an inadequate amount of NO₂, the Pt containing DPF can produce NO₂ from NO and improve the burn rate of the soot to a small degree. Increasing O₂ concentration in exhaust stream and holding the NO only concentration at the inlet led to an increase in NO₂ production for Pt containing samples. The formation of HNCO was observed over soot-loaded filters containing no washcoat. The HNCO formation resulted from the reaction of gas phase NH₃ and CO generated from the partial oxidation of soot. The presence of NO₂ assisted in increasing the formation HNCO at lower temperatures. One must account for CO, CO₂, and HNCO when calculating the regeneration rate of soot loaded filters containing no catalyst washcoat

INTRODUCTION

Due to increasingly stringent emission standards, coupled with customers' demand for better fuel economy, research pertaining to diesel engine and exhaust aftertreatment development is gaining a lot of attention. Diesel engines provide an improvement with respect to fuel economy but tend to also have increased levels of NO_x and particulate matter (PM) emissions compared to a gasoline vehicle equipped with a three way catalyst (TWC). Current strategies to control PM emissions utilize a diesel particulate filter (DPF) to filter the PM from the exhaust. Traditional filter development is performed on a vehicle or through the use of an engine dynamometer. For the purpose of evaluating a variety of DPF substrates and washcoat formulations, this process is not always practical due to cost and time constraints. Additionally, new filter materials are not always available as full-size parts. There is a critical need for a

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reliable, repeatable and efficient way of testing different DPF materials and formulations on a bench scale. A laboratory method for soot loading and regenerating DPF cores samples was used to study active and passive regeneration trends with reference to vehicle operating characteristics and on road use.

The primary approach to vehicle PM control uses a diesel particulate filter (DPF) that may be catalyzed (CDPF), which forces the exhaust gas flow through the substrate wall and removes the PM from the gas stream. More recent technologies that provide a potential to lower aftertreatment system costs incorporate a selective catalyst reduction (SCR) washcoat containing zeolite into the substrate for NO_x control and soot reduction simultaneously. This technology is sometimes known as a selective catalyst reduction filter (SCRF). Collected PM is combusted through the introduction of an energy source such as fuel to raise the temperature. Typical strategies use fuel injection across a diesel oxidation catalyst (DOC) to create an exothermic reaction. Fuel-borne catalysts have also been used to reduce soot oxidation temperatures [1]; however, non-combustible ash buildup in the filter remains a concern over the life of the vehicle. Advanced concepts regarding coating technologies for CDPFs has progressed and researchers have been able to show improvements to the soot oxidation rates by improving catalyst and geometry interaction [2]. Additional regeneration methods include use of microwave technology, electric heaters and stack scrubbers [3]. Such methods are used mainly for stationary industrial applications. There is little data demonstrating whether these methods would be cost effective or practical for vehicle applications. Many articles and studies have discussed the active and passive regeneration characteristics; our attempt is to understand and demonstrate the effects of various washcoat technologies under active and passive regeneration conditions using a bench scale flow reactor.

Typical vehicle DPFs separate carbonaceous gas-borne particles from the gas stream while the exhaust is allowed to pass through the porous substrate walls. For diesel applications, particles can be filtered from the exhaust gas stream through two types of filtration, depth filtration and cake filtration. Typically a cake filtration layer begins to develop under higher soot concentrations after the depth filtration mechanism has reached its saturation point. Depth filtration consists of three mechanisms of collection including diffusion deposition, inertial deposition and flow-line interception [4]. The DPF size, selection, and soot loading management are important to keep in mind so that backpressure of system does not drastically affect fuel economy and engine performance. The most common method of performing the regeneration process on vehicles is to use in-cylinder post-injection or downstream exhaust injection of diesel fuel, or combination of the two.

Basic strategy for active thermal regeneration, the DPF is loaded to a specified amount of PM. When regeneration begins, the carbonaceous portion of the PM is oxidized to yield CO or CO_2 . The desired product of PM combustion is CO_2 as opposed to releasing carbon black, because some studies have indicated that releasing the latter can be carcinogenic [5]. CO production from DPFs is considered to be the product of incomplete PM combustion. The rate at which PM or soot oxidization can take place is limited by kinetics and is the primary reason why soot cannot be effectively oxidized using a flow through catalyst technology such as those used for HC and CO. HC and CO oxidation occurs at a faster rate and can be accomplished in a flow through device. Since PM requires more time to combust, trapping is the only means that provides the increased residence time required for oxidation to occur. However, trapped particles will not combust without adequate temperature and O_2 concentration. If aftertreatment and engine conditions are desirable specifically NO_2 concentrations and catalyst inlet temperatures, regenerations can be performed passively.

There are two primary ways to limit the soot oxidation rate of a DPF: 1) temperature and 2) reactant concentration (O_2 and carbon). A temperature greater than 600°C allows the reaction to take place rapidly and completely whereas regeneration events performed at temperatures around 400°C are drastically slower and incomplete [6]. During a regeneration event under ideal conditions, particularly during the first few seconds, the oxidation rate is extremely fast, but as the mass fraction of soot decreases so does the reaction rate. Therefore, a more heavily loaded DPF would tend to regenerate at a faster rate than one that is lightly loaded. There is a limit to safe DPF loading and regeneration levels. Beyond this loading limit, regeneration can lead to uncontrolled heating that will deactivate or damage the DPF, in our experience such failures include deactivated washcoat, ring-off cracks and even melted sections of the substrate. The primary drawback to using an active regeneration strategy is the fuel economy penalty. A system that continuously regenerates the DPF is desirable (passive regeneration). Favorable conditions include a customer's drive cycle that provides exhaust gas temperatures of at least 300°C the majority of the time and a fairly high NO_x/PM ratio [7]. Having higher levels of NO_2 , at 50% of the NO_x or greater, allows passive regeneration to take place continuously. In order to create higher levels of NO_2 , an oxidation catalyst that has the ability to oxidize NO to NO_2 is required upstream of the DPF. However, for vehicle applications adding an additional catalyst may increase cost to the customer. Therefore, OEMs are attempting to use a combination of passive and active regeneration in their vehicle strategies to balance PM control and cost.

The main challenges faced when implementing passive regeneration on a vehicle has to do with the NO_2 concentration, or more simply put, there needs to be an adequate NO_2 concentration available for the process to take place. The other challenge relates to the substrate being in a temperature range that promotes passive regeneration. The temperature range for passive regeneration to take place requires the inlet temperature to the DPF to be in the 300°C 500°C range. Fully using the passive regeneration process can minimize the fuel economy penalty observed during active regeneration. One system that has been developed and has shown the ability use the passive function is the continuously regenerating trap (CRT) [8]. The CRT and other systems that have attempted to use this low temperature regeneration typically use a two stage passive diesel particulate filter system, where a non-catalyzed filter is regenerated downstream of an oxidation catalyst, which has the ability to produce sufficient amounts of NO_2 . In order for such passive systems to work properly a low sulfur fuel is required in conjunction with a high loaded Pt catalyst. By creating a sufficient amount of NO_2 in the exhaust stream, it is possible to regenerate the soot at temperatures around 300°C .

Passive regeneration systems in theory appear to be a very attractive approach to soot regeneration; however it is highly dependent on the engine duty cycle. If sulfur is kept out of the system you have addressed one of the main issues. However, at low load and idle conditions the temperature and NO_2 concentration that are required are difficult to obtain. If such low load or idle conditions are sustained for extended periods of time it may cause plugging of the CDPF and vehicle drivability issues. Thus, it would be ideal to have a system that is capable of both, leveraging each regeneration mode to keep cost impacts low. At this point, a major breakthrough in engine and aftertreatment technology combined would have to be made in order for passive to be a permanent solution to soot regeneration, however, we feel it is still important to understand the differences and see if there are areas where passive regeneration can assist in making the active regeneration interval longer. Low temperature conditions certainly exist in diesel passenger cars and other light-duty applications.

EXPERIMENTAL SETUP

For regeneration experiments, two different experimental test benches were utilized. The first is a soot generator which generates PM from the combustion of propane. Second is the regeneration reactor, which is used to regenerate PM loaded DPF samples.

(1) Samples were loaded using a Ford developed soot generation reactor, which generates and deposits the PM on the DPF sample. The soot generator combusts propane in an

O_2 and N_2 atmosphere to generate an optimized flame to create PM. Loadings are performed at a space velocity (SV) of 60k hr^{-1} . The generator includes a conditioning tunnel, which allows particles to coagulate to the correct size distribution to represent engine generated PM. The details of the soot generator loading process were previously discussed [9]. Figure 1 shows the particle size data taken from the soot generator and compared to two steady state vehicle operating points. The results illustrate the soot generator produces particles in the range of particles observed on the vehicle.

An extensive study was performed to validate the laboratory generated soot with engine generated soot. Loading as well as regeneration characteristics between laboratory and engine soot were quantified by authors in [10, 11]. Soot generated in the laboratory and from an engine showed good correlation with respect to soot loading and regeneration characteristics. Each of the different DPF samples cores were loaded to a target backpressure. The backpressure requirement was derived from targets found by loading a production sample to its target level.

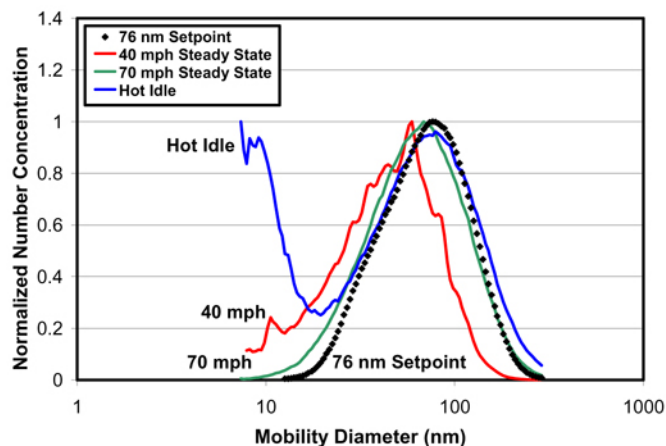


Figure 1. Particle size data taken from the soot generator and two steady state points (40 and 70 mph) comparing the soot generator's size distribution to vehicular exhaust PM size distributions. [9]

(2) In designing this regeneration reactor, several versions of the reactor bench were tested until settling on a design that was capable, effective and repeatable for DPF regenerations. Figure 2 shows the overall reactor layout for the regeneration reactor, which was used for passive and active regeneration experiments.

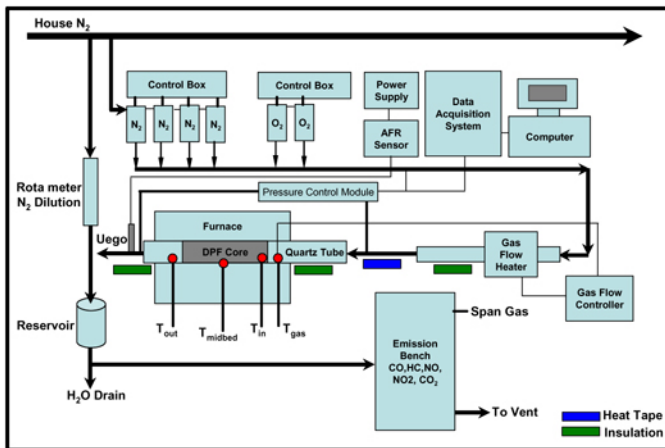


Figure 2. Regeneration reactor setup

The regeneration reactor can accommodate 1" diameter DPF cores of lengths ranging from 1" to 12" for plugged samples or flow through samples. This study used 3" cores for all regeneration validation experiments. This size was chosen to ensure that the internal flow characteristics are not compromised compared to standard aftertreatment components. DPF cores were wrapped in 1/16" glass fiber insulation and placed in an approximate 1-1/16" diameter quartz tube. At the exit of the reactor, N₂ gas is used to dilute the sample (2:1) before transfer to the emissions bench. A rotameter was used to control the dilution N₂ flow and allow for adjustment of the flow rate to a 2:1 ratio. Mixing of dilution N₂ and reactor exhaust is accomplished in a stainless steel ballast tank. Inlet and mid-bed thermocouples were installed from the outlet end of the DPF core. Once the sample is loaded into the quartz tube the appropriate flow gases, such as N₂, O₂, NO and NO₂ are switched on and a baseline backpressure is recorded. The temperature is ramped to provide a target T_{inlet} temperature. Depending on whether the regeneration test was passive or active such temperatures were different.

Back pressure was measured during the regeneration cycle by a 0 to 5 psi differential pressure sensor. The sensor was connected immediately upstream and downstream of the DPF core to obtain the pressure drop across the DPF core. Omega type K thermocouples were used to monitor the DPF core inlet, mid-bed, and outlet temperatures during regeneration. In addition, a thermocouple was placed 1" upstream and downstream of the DPF core to monitor the inlet and outlet gas temperature from the heater. A gas flow heater was used in addition to the furnace to create a more appropriate environment where inlet gas temperature is the key element in heating up the same. The furnace used around the sample is more for insulation and fine tuning of the temperature profile.

All samples tested in the regeneration test bench were loaded using a soot generator with particles ranging from 10nm to around 200nm. Samples were loaded to a pre defined backpressure to compare a variety of different DPF cores samples to our current production backpressure loading targets. PM loading was determined from measurements of the hot weight at 150°C of the DPF core before and after the PM loading procedure.

Table 1 below shows the set of experiments and various conditions used which included type of regeneration, NO and NO₂ ratios, O₂ concentrations and associated inlet temperatures.

Table 1. Experimental Test plan for active and passive regenerations.

T Inlet	O ₂	NO ₂ :NO _x	Regeneration Type
(°C)	(%Vol)	(ratio)	(.)
375	5	0.15	Passive
600	5	0.15	Active
600	2	0.15	Active
475	5	0.15	Passive
375	5	0.50	Passive

Following baseline pressure drop testing, the temperatures of the furnace and gas flow heater are simultaneously increased to provide the target inlet gas temperature shown in Table 1. There are two heating sources used to ramp the temperature, a heat gun to heat the inlet flow gases and a furnace which helps get the sample to the target regeneration temperatures more quickly. The heat gun provides the sample core with an environment that is more applicable to real world regenerations. Once the sample reaches the appropriate regeneration temperature, it is held there until the regeneration is close to 100% complete. The active regenerations are complete within 20 minutes where the low temperature passive regenerations takes significantly more time, sometimes several hours.

All regeneration experiments are allowed ample time for the regeneration process to occur to ensure that the mass on the DPF core has been fully regenerated or at least a sufficient amount of time to determine the associated soot burn rate. To quantify the completeness of the regeneration, the cores are removed from the quartz tube and placed in the furnace for one hour and baked at 150°C. Following the one hour baking, the DPF core is weighed to verify the core has returned to within an acceptable range of the initial clean weight, if not the weight difference is recorded. The PM fraction remaining on the DPF core following the regeneration process provides a computational means to evaluate the completeness of the regeneration. Through integration of the CO and CO₂ response curves it is possible to do a relatively accurate mass balance. A MIDAC FTIR was used to monitor the emission before, during, and after each of the different regeneration

experiments. Standards were generated for NO, NO₂, CO, CO₂, H₂O, and a variety of other compounds that are potential products during regeneration experiments. Emissions of CO and CO₂ were also measured with a secondary set of analyzers as a back up to the FTIR.

The SV during the different regeneration experiments was approximately 60k hr⁻¹ and exhaust flow rates were around 22 L/min. This space velocity was chosen to approximate the average SV derived from the typical mid-range vehicle space velocity range derived from Federal Test Procedure (FTP) testing. The pressure drop results showed very similar profiles with each sample achieving a slightly different peak pressure drop; however the duration to achieve the target loading was different as a result of different washcoat formulations, or initial backpressure differences. The small variability in peak pressure values is a result of minor soot loading differences.

To gain understanding of the passive and active regeneration characteristics, a variety of different samples were used during laboratory test regenerations. All test formulations were done on the standard SD021 SiC 200/18 substrate with 42% porosity. There was a blank, low Pt only, low Pd only, Cu/zeolite, Pt:Pd 1:1, and a high Pt only sample. The samples that were used in all regeneration experiments are shown in Table 2.

Table 2. Sample information for passive and active regeneration experiments.

Sample ID	Washcoat Loading
	g/ft ³ (Pt:Pd)
A	0 (0:0) Blank
B	5 (1:0)
C	5 (0:1)
D	Cu/Zeolite
E	5 (1:1)
F	30 (1:0)

All core samples were degreened in a furnace at 650°C for four hours, followed by a single regeneration where the samples are loaded to their maximum soot loading.

Finally, results obtained from the laboratory regeneration experiments were applied to a couple of different steady state points to determine if passive rates were reliable and to what extent passive opportunities could be used for real world applications. The engine used to gather steady state data points was a 6.4 L V-8 diesel engine equipped with a DOC and DPF. Horiba test benches and MKS FTIRs were used to measure emissions values such as NO and NO₂. A real time mass measurement system was used to determine the soot loading rates on the CDPF component. Omega K

thermocouples were used to measure the temperatures in the exhaust stream such as the inlet temperature to the CDPF.

RESULTS AND DISCUSSION

Each of the different DPF sample cores were loaded to the target backpressure. Sample B is a current production sample and was used to develop the target level. To establish our loading criteria it was soot loaded to approximately 5.5 g/L where the pressure drop across the DPF sample was recorded and used as the loading criteria for loading and testing all the samples from A through F. If necessary, additional soot loading levels were performed and regenerated. Each of the DPF samples were loaded to the target pressure and the loading curves for A - F samples are shown in Figure 3.

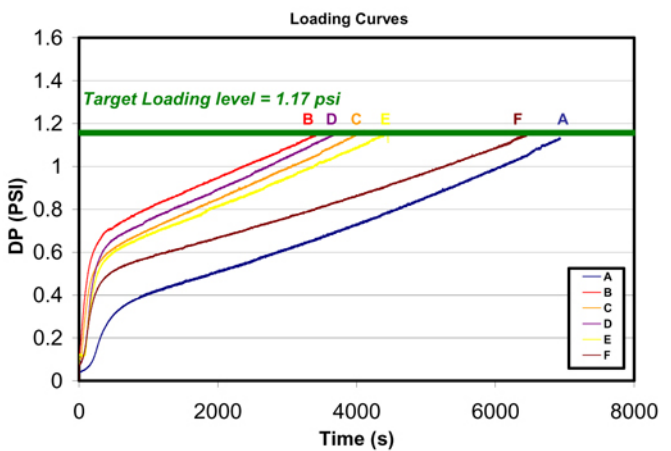


Figure 3. Loading curves for DPF samples A, B, C, D, E, and F.

As shown in Figure 3, as the loading time to get to the target back pressure increases so does the soot loading level. This phenomenon is expected because as washcoat levels decrease the initial loading phase within the DPF wall is slower, as more mass is required to reach the target back pressure. Table 3 shows the Loading levels for each of the different cores samples A - F.

Table 3. Samples information for passive and active regeneration experiments.

Sample	Soot wt (g)	Loading (g/L)
A	0.2914	10.1
B	0.1575	5.4
C	0.1832	6.3
D	0.1590	5.5
E	0.2091	7.2
F	0.2787	9.6

Note that for all the different active and passive regeneration experiments the soot loading was stopped when samples reached the target back pressure of 1.17 psi. In addition, an