

Degradation of Nonmethane Hydrocarbon Oxidation Efficiency of a Catalyzed Diesel Particulate Filter during Aging

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ABSTRACT

Upcoming 2013 on-board diagnostic (OBD) requirements mandate that catalyzed diesel particulate filter (CDPF) non-methane hydrocarbon (NMHC) oxidation efficiency degradation be monitored as the filter ages. The malfunction indicator lamp (MIL) is to light when exhaust NMHC levels are greater than two times the legal limit or when the CDPF has no detectable amount of NMHC conversion capability. In this study, a CDPF was thermoelectrically aged at 650°C from the degreened (near-new) to end of useful life (EUL) stage. CDPF total hydrocarbon (THC) oxidation efficiency was measured periodically using a suite of steady-state speed and load conditions ("snapshot test"). The snapshot test was performed after every seven hours of filter aging. Snapshot test data of steady-state THC oxidation efficiency change across the filter at the chosen operating conditions shows no discernible, consistent degradation as the filter is aged. When uncertainty analysis is applied to the results, the story is even clearer: It is impossible to tell, with statistical significance, the age of a filter from THC oxidation efficiency using this methodology. Light-off shift - the gradual upward shift in light-off temperature due to advanced catalyst age - shows promise as a potential method for determining CDPF age with respect to NMHC oxidation efficiency. Light-off temperatures of degreened and EUL filters are separated by more than 50°C. Using this temperature shift as a diagnostic tool has already been successfully applied to diesel oxidation catalysts (DOCs), but will require different strategies and detection algorithms to apply to CDPFs.

INTRODUCTION

CDPFs, which are responsible for collecting the soot that is an intrinsic product of diesel combustion, are a necessary component of modern diesel aftertreatment systems. CDPFs, however, also affect other exhaust constituents in addition to filtering soot. Although NMHC is the exhaust constituent of interest from a regulatory standpoint, THC was measured in this study since THC is representative of NMHC levels and is able to be directly measured (whereas NMHC concentration is a derived quantity using steady-state measurements of THC and CH₄). The exhaust constituent of interest to this study is gaseous THC, which is oxidized across the CDPF into CO₂ and H₂O. This paper is not concerned with particulate emissions.

OPERATING PRINCIPLE

A CDPF is a porous ceramic monolith generally made from materials like cordierite or SiC with a washcoating of precious metals (commonly a mixture of Pt and Pd) [3]. The CDPF used in this study is a wall-flow filter. This filter has alternating blocked channels which force the exhaust flow through the ceramic wall of the filter. Most exhaust gases are able to pass through the filter, but larger soot particles are not and remain physically trapped on the substrate's surface, and therefore are prevented from venting through the tailpipe (see [Figure 1](#)). A mechanically-healthy soot filter is capable of filtering 99% of soot from the exhaust stream.

Eventually, the filter will fill with soot and require cleaning, a process referred to as "regeneration" [3]. Active regeneration

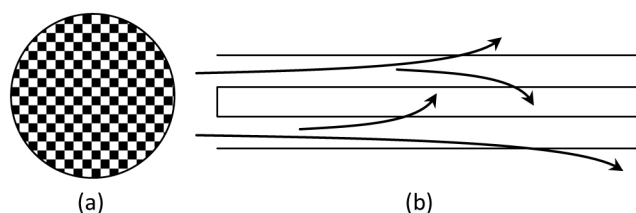


Figure 1. Cross-sectional view of CDPF showing blocked channels in the front view (a) and exhaust wall flow in the interior projection (b)

involves heating the filter above the soot's combustion temperature, which is nominally higher than 600°C, and much higher than typical diesel engine exhaust temperatures [2]. The introduction of the catalyst washcoat, however, lowers this soot combustion temperature significantly by allowing regeneration at nominal engine-out exhaust temperatures in the presence of NO₂ (which is referred to as *passive regeneration*) [3]. Since fuel is spent to heat the filter, the lower the ignition temperature of the soot, the less fuel required, and therefore the lower the fuel penalty (i.e., the additional fuel used to heat the aftertreatment that is not used for power generation).

AGING MECHANISMS

While CDPFs have a multitude of physically-based failure modes (cracking due to excessive pressure drop, cracking due to thermal stresses, melting due to runaway regeneration, separation of the monolith from the can), these are discrete failure modes [5, 6]. The catalyst washcoat, however, degrades over time as the filter is used. The performance of a CDPF's catalyst can degrade due to soot loading level, changing geometry of the filter's substrate, the sulfur common in diesel fuel (despite the lower levels since 2007), and heat. Catalyst fatigue is commonly referred to as "aging."

A high soot load on the filter degrades catalyst performance through physical blocking of possible reaction sites. Filtered soot can be deposited on top of catalyst molecules, which prevents the catalyst from reacting with passing exhaust. This degradation is alleviated by regeneration. Catalyst reaction sites can also be blocked by deposits of incombustible ash which is a byproduct of the combustion of lubrication oil. Ash is not removed from the filter by the regeneration process.

Changing CDPF geometry on the microscopic scale is another cause of catalyst degradation. As the filter is heated and cooled, the pores in the filter expand and shrink. During this thermal cycling, pores can collapse, trapping the catalyst washcoat that was present in that pore. This is an irreversible form of aging.

In addition to hydrocarbons, diesel fuel contains many chemicals that result from the drilling and refining process. These chemicals, especially sulfur, react with the catalyst and poison the precious metals, eventually rendering microscopic regions of the catalyst inactive. Ultra Low Sulfur Diesel (ULSD) diesel fuel helps limit this reaction and slow this particular method of aging.

Heat is, above all other aging modes, the most detrimental to catalyst health. Under nominal engine-out heat levels, the catalyst on a CDPF would easily last longer than the vehicle itself. It is the extreme heat generated during active regeneration that causes damage by sintering the catalyst atoms deposited on the surface of the substrate. This sintering results in a measurable loss of catalyst surface area, and therefore, a loss in overall reactivity with soot [4].

REGULATION

Although the catalyst was originally designed to reduce the temperature at which soot oxidizes during regeneration, new regulations require the monitoring of the catalyst's health with respect to alternate diesel exhaust emission constituents. On-board diagnostic (OBD) requirements for 2013 require that the engine computer be able to diagnose a CDPF that has NMHC emissions greater than 200% of the OBD threshold, or if the catalyst is no longer capable of converting NMHC [1]. The diesel engine used in this study, especially with the inclusion of the upstream oxidation catalyst (see Figure 2), will not exceed the 200% threshold. Thus, the aim of this study is to ascertain when the CDPF has no hydrocarbon oxidation capability.

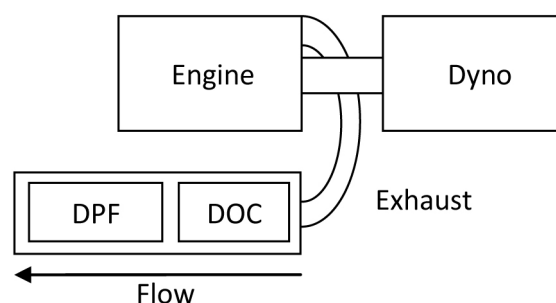


Figure 2. CDPF experimental setup

SNAPSHOT TESTING

TEST PURPOSE

In order to better-understand the severity of CDPF NMHC oxidation efficiency degradation, the filter was subjected to a series of aging and testing cycles. These cycles, the so-called "snapshot tests," were designed to characterize the NMHC oxidation efficiency of the filter as it aged from degreened to EUL in a reliable, highly repeatable manner. The snapshot test was also performed on a fully-inert filter to discern the

difference between an EUL filter (which should still pass emissions testing) and a CDPF with a catastrophically failed catalyst.

Table 1. Aftertreatment Specifications (* denotes IP restrictions)

	Parameter	Actual Performance
DOC	Height	5 in. (monolith)
	Diameter	9.75 in. (monolith)
	Configuration	Flow-Through
	Substrate Material	Cordierite
	Catalyst Material	Precious Metals Washcoat*
CDPF	Height	11.25 in. (monolith)
	Diameter	9.75 in. (monolith)
	Configuration	Wall-Flow
	Substrate Material	Cordierite
	Catalyst Material	Precious Metal Washcoat*

AGING PROCEDURE

The filter was thermally aged in an electric furnace normally used for annealing metals. Thermoelectric aging was chosen instead of on-engine aging in order to achieve a repeatable process that is consistent with industrial practices. The procedure for aging is detailed in [Table 2](#). This aging procedure alternated with the snapshot cycle detailed in [Table 3](#). Even though each aging segment took 24 hours, the filter was only aged for seven hours at 650°C; filter age is reported in seven-hour increments.

Table 2. Aging Procedure

Time (hours)	Temperature (°C)
00:00	25
02:00	650
09:00	650
24:00	25

EXPERIMENTAL APPARATUS

The snapshot test was performed on a preproduction 2007 Cummins 6.7L ISB. Engine speed and torque were automated through a dSPACE data acquisition system to increase test-to-test repeatability.

The measurements were conducted in the enclosure of the aftertreatment system. Temperature and pressure were measured with the standard production sensor harness that is nominally installed with this system in any field application. [Figure 3](#) illustrates the location of the aftertreatment instrumentation.

Emissions constituents were measured by analyzers provided by California Analytical Instruments (CAI). CO₂ was not measured since the aftertreatment system accumulates CO₂ (in the form of uncombusted soot), which would introduce a

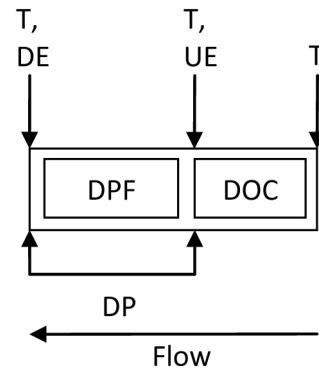


Figure 3. Measurement Locations: T = Temperature, DP = delta pressure, UE = upstream emissions, DE = downstream emissions

difficult-to-reproduce generation term into the measurements. The NO/NO₂ split was not measured for this experiment due to available analyzer limitations. Upstream and downstream measurements were taken by the same set of analyzers using an automated switching valve.

TEST PROCEDURE

Individual snapshot tests were performed between aging cycles to characterize the degradation in CDPF THC oxidation efficiency as the filter aged. These tests consisted of seven steady-state speed and torque points which are outlined in [Table 3](#).

Table 3. Snapshot test procedure

	Time (s)	Speed (rpm)	Torque (lb-ft)
Test	0 - 930	760	0
	946 - 1572	1500	150
	1584 - 2214	2000	350
	2226 - 2856	2200	175
	2868 - 3498	2500	400
	3510 - 4380	1800	100
	4392 - 5022	2500	625
Cooldown	5034 - 5154	1800	100
	5166 - 5466	760	0

The points selected for the snapshot test (see [Table 3](#)) are based on the Environmental Protection Agency's (EPA's) 13-mode steady-state test points. The order of the test points was chosen to include a number of operating conditions namely: engine warmup (760 rpm / 0 lb-ft), gradual increase in