

In-Situ Optical Analysis of Ash Formation and Transport in Diesel Particulate Filters During Active and Passive DPF Regeneration Processes

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ABSTRACT

The formation and transport processes governing the build-up of incombustible ash deposits in diesel particulate filters (DPF) are influenced to a large extent by the filter's operating history. More specifically, the regeneration process, whether active, passive, or some variation of the two, has long been assumed to exert significant influence on the resulting ash characteristics. Until recently, only limited circumstantial evidence was available to describe differences in ash properties and distribution impacting DPF performance for filters subjected to different regeneration strategies. This work presents, for the first time, results from a comprehensive series of evaluations with optically-accessible DPF core samples showing the processes controlling the formation, transport, and interaction of the soot and ash deposits over a range of DPF regeneration conditions.

DPF sample fixtures with optical access were employed with core samples from new and aged (ash-loaded) catalyzed DPFs to visualize the soot oxidation and ash formation processes occurring during filter regeneration. The filter core samples were loaded with soot utilizing a small diesel engine. Regeneration studies were carried out on a bench reactor allowing for careful control of exhaust conditions (temperature, flow, and gas composition). A stereo microscope provided high resolution videos of the processes occurring during the regeneration event in a small portion of a single DPF channel. Measurements of filter pressure drop and gaseous emissions via FTIR provide additional data, correlated to the microscope videos, to quantify the soot oxidation processes.

Results of this study clearly show strong interactions between the ash and soot during the regeneration event. In particular, soot oxidation in the catalyzed DPF is observed to proceed locally with a certain degree of mobility of the soot cake as it is oxidized, depending on the specific regeneration pathway. Regeneration with relatively thick soot cake layers further shows the manner in which ash precursors are concentrated in the remaining soot and ultimately begin to agglomerate and form larger ash particles in the DPF. Particle mobility and transport of ash and soot from the channel walls to the back of the DPF were also explored during the regeneration event and following partial regenerations. The results shed considerable light on the processes occurring in the DPF and are extremely useful to describe the physical mechanisms governing ash formation and transport in the DPF, and ultimately explain the impact of ash on DPF performance.

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INTRODUCTION

Particulate filters, now ubiquitous for most diesel applications, have recently attracted increasing attention as a potential means for reducing particulate matter (PM) emissions from gasoline direct injection engines as well. The widespread application of these filters is due in large part to their high degree of effectiveness for trapping particles emitted from the engine. Although the vast majority,

generally over 99% by mass, of particles emitted from modern diesel engines consists of soot, defined as combustible material composed of carbon, sulfate compounds, and other organic matter, the remaining 1% of particle emissions consists of ash [1]. On the other hand, the relative proportion of ash-to-soot emitted from gasoline direct injection engines is expected to be greater, due to the relatively low soot mass emissions compared to oil consumption [2].

Unlike soot which is oxidized during regeneration of the diesel particulate filter, the incombustible ash, composed primarily of metal oxides, sulfates, and phosphates, remains. While the majority of the ash originates in the lubricant additives, calcium- and magnesium-based detergents, and zinc dialkyldithiophosphate (ZDDP), additional sources of ash include engine wear metals, corrosion products, and trace metals in diesel fuels [3]. Over time, the accumulation of ash in the particulate filter restricts exhaust flow through the filter, decreases the available filtration area, negatively impacts fuel economy, and ultimately determines the service life of the filter.

Impact on DPF Performance

The deleterious impact of ash on DPF flow restriction and pressure drop are well-known. Previous studies have shown an increase in filter pressure drop in the range of 50% to 100% due to the ash alone [4,5,6]. Considering the compounding effect of soot accumulation in a DPF with a moderate to high level of ash, the increase in pressure drop over the clean filter can be even greater, due to an increase in the DPF's pressure drop sensitivity [5, 7, 8]. Overall, estimates show a potential increase in fuel consumption, due to increased exhaust flow restriction and regeneration frequency at elevated filter ash levels, in the neighborhood of 1% to 3% [7].

While the impacts of elevated ash levels on DPF and vehicle performance, outlined above, are well-understood, more recent studies point to more subtle, yet beneficial effects, at low ash loads during the initial stages of the DPF “break in” and ash build-up process. In particular, the accumulation of a small amount of ash (< 10 g/L) has previously been reported to form a thin layer or membrane along the filter walls, preventing soot depth filtration and resulting in a pressure drop reduction relative to a clean DPF (with no ash) for the same level of soot accumulation [4, 5, 9]. Additional work at MIT, Figure 1(a), and the University of Wisconsin have shown that even extremely low ash-levels, less than 2 - 3 g/L can contribute to a significant reduction in the soot-related pressure drop [10].

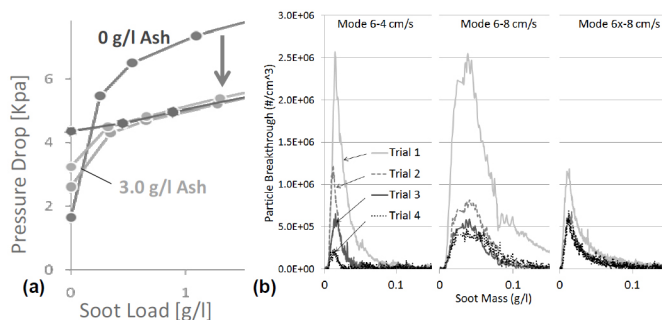


Figure 1. Impact of ash membrane to reduce filter pressure drop (a) and particle break through (b) [10].

The data shown in Figure 1(a), from work investigating the impact of the initial ash build-up process on DPF

performance, indicates a nearly 33% reduction in DPF pressure drop with only 3 g/L of ash accumulation. Additional studies utilizing DPF wafers have shown similar results with less than 2 g/L of ash accumulation [10]. Both of these studies indicate that even at these low ash levels, the ash sufficiently covers the DPF surface pores (no significant penetration deep into the substrate was observed) to considerably reduce the exposed surface pore area available for soot depth filtration.

Figure 1(b) shows the measured DPF particle breakthrough from the University of Wisconsin study [10]. Each trial represents the accumulation of approximately 0.25 g/L ash on the DPF wafer, and shows over an 80% reduction in particle breakthrough with the accumulation of approximately 2 g/L ash on the filter surface, relative to the clean wafer with no ash. Even the accumulation of 0.25 g/L ash after only Trial 1 in this study showed a reduction in particle breakthrough of over 50% for nearly all flow conditions [10]. Therefore, while a complete ash layer or membrane has not yet fully-developed at these low ash loads, the accumulation of ash in (or over) the filter surface pores provides a considerable improvement in particle number trapping efficiency.

Ash accumulation in the DPF, thus, presents both potential advantages and disadvantages from an overall filter performance and engine efficiency perspective, depending on the amount and location of the ash. In order to better understand these, at times, competing processes and exploit them to mitigate the ash problem, the details of the initial stages of the ash build-up process and subsequent evolution of the ash deposits must be better understood.

Ash Emissions Morphology

In order to understand the ash formation and transport processes occurring within the DPF, it is first necessary to appreciate the origin and morphology of the ash emitted from the engine and entering the DPF. Figure 2(a) presents a TEM image from work at the Tokyo Institute of Technology showing a small, 10 nm, calcium ash precursor bound to an engine-out soot particle [11].

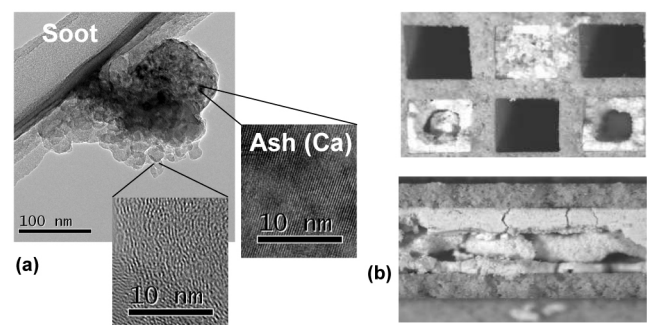


Figure 2. Ash precursors bound to soot agglomerate in the exhaust (a) [11] and ash accumulated in DPF (b) [5].

Previous studies have reported lubricant-derived ash-related elements, including calcium, zinc, magnesium, and phosphorous to be emitted from the engine in small, less than 100 nm sized particles, bound to the carbonaceous soot. In particular, a 2007 study collected individual engine-out soot particles from the exhaust of a Cummins ISB engine and found all of the lubricant-derived elements bound to the carbonaceous soot, whereas engine wear and corrosion particles were found to exist in the exhaust as distinctly separate particles [1]. The TEM images presented in [Figure 2\(a\)](#) are further consistent with these findings.

More recently, additional studies by Mayer et al. have reported similar findings, identifying significant metal oxide nano-particle emissions in the 10-30 nm size range. In particular, studies with ceria-based fuel-borne additives showed nearly all of the ceria emitted from the engine bound to the carbonaceous soot (below a threshold ceria dosage rate). Above the threshold ceria consumption rate, cerium oxide particles in the 10-30 nm size range were detected to exist as distinctly separate particles from the carbonaceous soot [12]. These results imply that below a certain ash-to-soot emissions ratio, the nano-particle size ash precursors are primarily emitted bound to the carbonaceous soot. However, when the ash emissions (either via excessive oil consumption or fuel-borne catalysts addition) become sufficiently high relative to the soot emissions, the ash may be emitted as distinctly separate particles.

This theory is further supported by additional work measuring the composition of size-selected soot emissions via inductively coupled plasma (ICP), which showed elevated lubricant-derived metallic concentrations in the smallest particle size fraction, below 30 nm. Over the majority of the engine operating conditions evaluated, the metal emissions appeared to be bound to the carbonaceous soot, with the exception being the engine idle condition where the oil consumption remained relatively high in comparison to the soot emissions. Interestingly, the majority of the zinc particles were measured in the soot fraction collected below 30 nm, whereas the calcium particles were detected in roughly equal amounts throughout all of the particle size fractions [12,13,14]. While the reason for these differences is unclear and additional work in this area is required, these studies provide further evidence that the majority of the engine-out ash is emitted as very small, nanometer-scale particles.

The specific ash consumption mechanism in the engine most likely controls the size and morphology of the engine-out ash. In the case of fuel-borne catalyst emissions measured by Mayer, the catalyst experiences the full combustion process in the cylinder along with the diesel fuel. Similarly, much of the lubricating oil which makes its way through the piston ring pack and top ring zone to the combustion chamber also experiences the combustion process and is emitted as nano-size particles bound to the carbonaceous soot, as reported in [1, 11, 12]. On the other hand, ash emissions in the form of liquid (un-combusted) oil, such as through valve

or turbocharger seal leakage, may exhibit differences in particle morphology from ash which has experienced the combustion process. A recent study published by Daimler, highlighted the potential for different oil consumption mechanisms to impact the size of the resulting ash particles, with liquid oil consumption leading to larger particles relative to ash which had experienced the in-cylinder combustion process [6]. Previous work by Zarvalis et al. simulating liquid oil consumption and its impact on ash emissions, showed similar results, with the formation of larger ash particles in the exhaust [15].

Although there does appear to be a correlation between oil consumption (or ash consumption) mechanisms and the resulting ash particle size in the exhaust, the majority of the work to date indicates the size of the engine-out ash to be extremely small, certainly less than 100 nm, and bound to the carbonaceous soot under most operating conditions. The notable exception to this observation being engine idle, which in some cases can be a large fraction (non-negligible) of the overall vehicle operation.

Despite the ash being emitted from the engine as relatively small particles, analysis of the ash deposits in the DPF, [Figure 2\(b\)](#), show the presence of much larger, micron-size ash agglomerates. Measurements of particle size for ash deposits found in the DPF report average particle sizes typically ranging from 1 - 10 μm and up to 200 μm in some cases [4,5,6, 16]. This represents an incredible increase in ash particle size by over several orders of magnitude in the DPF. Clearly the ash formation and agglomeration processes occurring in the particulate filter play an important role in the size and properties of the resulting ash particles.

Ash Formation and Transport

It is the objective of this work to investigate the mechanisms governing ash accumulation and subsequent particle agglomeration, growth, and transport in the DPF. The properties of the resulting ash agglomerates, as well as the location of the ash deposits, directly impact DPF performance. Understanding these fundamental processes is, thus, a crucial first step to developing means to mitigate the ash problem.

Since the ash agglomerates are formed from the nano-size ash precursors in the DPF directly, over tens of thousands of miles and several thousand hours, it is not unreasonable to assume that the operating history of the DPF over the course of this long residence time may play a role in influencing ash formation and distribution in the particulate filter. In fact, a review of the literature indicates a small, but growing, body of circumstantial evidence pointing to differences in regeneration conditions and the resulting ash properties. However, as this study will show, it is not necessarily the type of regeneration, whether active or passive, that exerts the greatest influence on the ash properties, but more specifically a small subset of parameters which are often difficult to decouple from the overall regeneration process as a whole.

Nonetheless, it is instructive to first examine the various reports in the literature to frame the context of this discussion. From the small body of literature reporting on differences in regeneration strategy, as they relate to ash accumulation and DPF performance, the majority report passively regenerated filters resulting in more ash deposited in a layer along the channel wall and less ash deposited in the end plug. On the other hand, actively regenerated filters tend to show much less ash deposition on the channel walls and more ash packed at the end of the DPF channels [4, 17, 18]. More recent work has further indicated the quality of the regeneration process (whether or not all of the soot is completely oxidized), the frequency of the regeneration, and the origin and size of the ash particles may be more important than whether the DPF is actively or passively regenerated in determining where most of the ash is deposited in the DPF [6]. In addition, the amount of soot at the start of regeneration and peak temperatures attained during regeneration have also been correlated to the resulting ash agglomerate size [18]. The sensitivity of the ash to variations in exhaust conditions, and ultimately the impact of the ash on DPF pressure drop, has also been linked to the ash chemistry and original lubricant composition [3].

The reported differences in ash distribution and ash properties governing the impact of the ash on exhaust flow restriction (permeability, porosity, packing density) are not trivial. A study by Dittler showed a passively regenerated DPF exhibiting 66% greater pressure drop than an actively regenerated DPF for the same ash level, with the differences in ash distribution attributed to the quality and frequency of the regenerations [6]. Work by Sappok et al. previously reported a continuously regenerated DPF exhibiting an increase in pressure drop of approximately 55% relative to a DPF that was regenerated periodically at the same ash level [4].

Computational studies have been less conclusive. Initial studies indicated optimal ash distribution to reduce total DPF pressure drop dependent on the loading state of the filter. For the case of ash alone (no soot), the lowest pressure drop was achieved by packing all of the ash in the end plug, which resulted in 5%-14% lower pressure drop than depositing all of the ash on the channel walls. The opposite effect was observed for the case of soot accumulation at 6g/L, where the lowest pressure drop was achieved by depositing all of the ash on the channels walls (25%-33% lower than packing the ash in the end plugs) [18]. More recent work has shown the effect of ash distribution to be more complex and highly sensitive to the specific ash properties (packing density and permeability) and composition, which may vary with lubricant chemistry and position in the DPF [19].

The difficulty in interpreting the broad range of observations reported in the literature lies in the fact that many of the underlying parameters controlling the ash formation and transport processes in the DPF are, to some extent, coupled to the specific regeneration process. For example, active regenerations are generally carried out after a

substantial amount of soot has accumulated in the DPF, and may occur under significantly different temperature, exhaust composition, and exhaust flow conditions than a passively regenerated DPF. On the other hand, a passively regenerated filter may operate with a little soot accumulation under a very different set of exhaust conditions. In order to identify the controlling processes, it is necessary to first decouple them through a series of carefully designed experiments.

EXPERIMENTAL PROCEDURES

The experiments described in this study utilized DPF core samples mounted in a specially designed fixture allowing for optical access to a small portion of a single DPF channel in order to visualize the soot oxidation, ash formation, and transport processes. All experiments were conducted with filter samples pre-loaded with soot and ash using a catalyst bench reactor to enable careful control of the regeneration conditions.

DPF Specifications

The DPF samples used in this study consisted of core samples removed from conventional D5.66" (14.38 cm) X 6" (15.24 cm) cordierite diesel particulate filters. The cell geometry of 200 cells/in.² (31 cells/cm²) and wall thickness of 0.012 in. (0.30mm) were the same for all of the samples, which were also coated with a platinum catalyst and washcoat.

The bench reactor testing utilized cylindrical cores 0.875" in diameter (2.22 cm) X 6" (15.24 cm) long. The small cylindrical cores were wrapped, using an intumescent mat mount, and canned in stainless steel sample holders. Use of the sample holder facilitates installation of the core samples on the flow bench for pressure drop measurements and performance characterization, while at the same time enabling optical access.

Soot loading of the core samples was accomplished by diverting a small portion of exhaust from either a 2002 Cummins ISB or 2009 Yanmar L100 to flow through the core sample fixture. The Cummins ISB, as used in this application, has been described extensively previously [4, 7, 20]. The Yanmar L100 is a 435cc, direct injection, air-cooled, single cylinder diesel. The engine is rated at 6.8 kW at 3,600rpm. In some cases, it was desired to increase the ash content of the soot, to facilitate visualization of the ash formation and transport processes in the DPF. In these cases, the ash content was enriched utilizing a burner-based accelerated ash loading system in conjunction with the Yanmar diesel engine.

The accelerated ash loading system has also been described in several previous publications in detail [4, 7, 20]. This system allows for control and acceleration of specific oil consumption mechanisms, enabling the simulation of engine-out ash and PM as closely as possible. Previous studies showed similar exhaust ash emissions characteristics between the accelerated ash loading system and Cummins engine [20]. Further, a comparison of DPFs aged in the laboratory using

the accelerated ash loading system with filters subjected to 180,000 miles of on-road fleet testing using CJ-4 oils showed ash properties, distribution, and composition to be very similar for both the laboratory and field-aged filters [4].

The enrichment of the ash content in the soot by means of the accelerated ash loading system was accomplished using a fully formulated, commercial CJ-4 oil. The oil contained 1,388 ppm Ca, 355 ppm Mg, and 1,226 ppm Zn, and the detailed composition of the oil can be found in [21]. Conventional ultra-low sulfur diesel (ULSD) was used in both the engine and burner system for all of the tests.

Optical System

The optically-accessible DPF core sample fixture is shown in Figure 3. The fixture enables optical access to only a small portion of the outermost filter channel. Figure 3(b) shows a cross-sectional view in which a segment of the top filter wall of the channel has been removed to expose the channel surface below. Use of a stereo microscope in conjunction with this fixture enables real-time imaging of the soot oxidation and ash formation process on the surface of the filter channel. An electrical heater (not shown) is used to minimize the heat losses near the viewing window. The current sample fixture provides a rectangular field of view approximately equivalent to the width of the channel (slightly more than 1 mm) and several millimeters long.

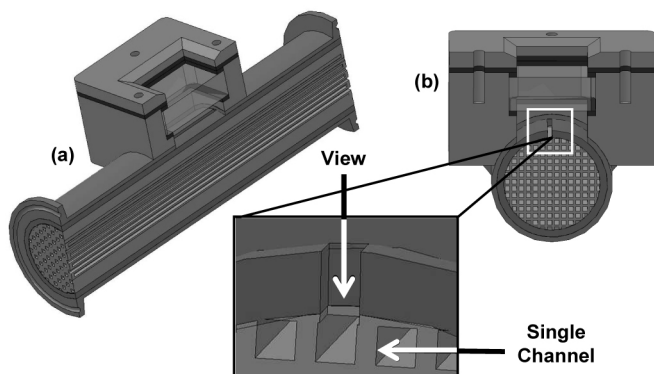


Figure 3. DPF sample fixture with optical access (a) and cross-section showing view into single channel (b).

The DPF core sample with optical access is designed to allow for soot loading on the engine, and regeneration on the catalyst bench reactor. The bench reactor, shown in Figure 4, enables control of inlet gas composition, flow and temperature, as well as measurements of total filter pressure drop and DPF temperatures. An MKS FTIR analyzer was also configured to monitor changes in gas composition upstream and downstream of the core sample during the regeneration process.

Unless otherwise specified in the discussion of the regeneration results, gas composition used with the bench reactor consisted of 500 ppm NO with dry air balance, as the objective of this study was to visualize the fundamental physical processes occurring during the regeneration event, as

opposed to understanding the catalyst performance. Further, soot loading at the beginning of the regeneration was around 4 g/L, resulting in a well-established soot cake.

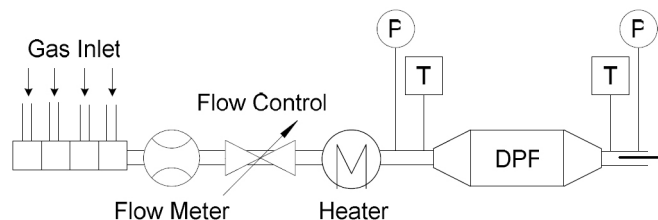


Figure 4. Catalyst bench reactor and associated measurement and control systems.

Figure 5 presents an example of a filter regeneration event with a well-established soot cake layer on a clean DPF (no ash). The regeneration was conducted at a constant temperature of 450 °C at 40,000 hr⁻¹ with the gas composition as noted above. The images have been shifted in time to account for a slight delay in the regeneration process occurring in the optically accessible channel at the periphery of the DPF. Despite the addition of heating elements to the window section of the sample fixture, some degree of heat losses in this region are unavoidable. The images and their associated timing with the pressure drop data, presented in Figure 5, should thus be interpreted in a qualitative sense.

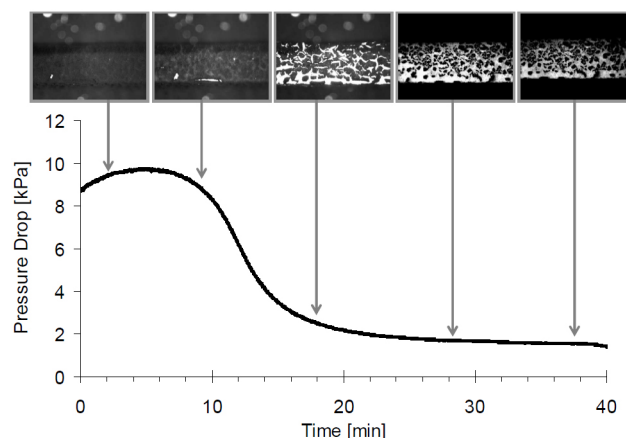


Figure 5. Pressure drop reduction and corresponding images of soot oxidation on DPF surface.

The application of the optical system to visualize the basic physical ash formation and transport processes in the DPF, provide a unique set of data, acquired under carefully-controlled conditions to investigate the impact of variations in regeneration and exhaust parameters on DPF performance.

RESULTS AND DISCUSSION

The presentation and discussion of the results that follow are organized under two general topics: (1) ash agglomerate formation, and (2) ash and soot agglomerate transport in the DPF. Interpretation of the experimental observations draws