

Theoretical and Experimental Analysis of Ash Accumulation and Mobility in Ceramic Exhaust Particulate Filters and Potential for Improved Ash Management

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ABSTRACT

Ash accumulation in the channels of ceramic, honeycomb-type particulate filters is controlled by several key parameters, which are the focus of this study. Ultimately, it is the formation of ash deposits, their transport, and the manner in which the ash accumulates in the particulate filter, which determines the useful service life of the filter and its resulting impact on engine performance. Although significant variations in ash deposit properties and their spatial distribution within the filter channels have been reported, depending on the filter's application, understanding the key parameters and mechanisms, such as the effects of exhaust flow and temperature conditions, as well as the processes occurring during filter regeneration events (whether passive or active) are critical in developing improved filter ash management strategies.

This work combines fundamental modeling studies with in-situ optical investigations clearly showing the processes whereby ash deposits are formed within the particulate filter, and subsequently transported down the length of the filter channel. A one-dimensional model was developed to describe the ash particle transport inside the filter channels, estimating the flow friction force imposed on the ash particles based on the local mean flow velocity. Size-dependent effects (critical mass) are accounted for in the model, which includes the re-entrainment of smaller, micron-sized ash particles, as well as the transport of larger agglomerated ash deposits (larger than 500 μm) through surface rolling or sliding. The results of the simplified 1-D model are compared and contrasted with experimental results from the in-situ optical studies, as well as three-dimensional analysis of the flow fields within particulate filter channels reported in the literature. Consistent with experimental observations, the theoretical analysis presented in this work provides a framework for understanding, and potentially controlling, the fundamental processes governing the mobility and accumulation of ash deposits within particulate filters, whether they are used as diesel particulate filters (DPF) or gasoline particulate filters (GPF).

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INTRODUCTION

Wall-flow particulate filters have seen widespread use to reduce particulate matter (PM) emissions from diesel sources for a large range of on- and off-road applications. Diesel particulate filters (DPF) function by trapping particles in the exhaust gas along the porous channel walls in a surface layer (cake filtration) and within the pores of the channel walls (depth filtration). The trapped particles, which consist primarily of a carbonaceous component, soluble organics, sulfates, and a small amount of ash, are subsequently oxidized to maintain PM levels in the filter below an acceptable threshold level. While the vast majority of the PM constituents are combustible, and are readily removed from the filter via oxidation (regeneration), a small amount of incombustible ash remains. In most cases, this incombustible material, which consists of metal oxides,

sulfates, and phosphates, is derived primarily from lubricant oil additives, however fuel-related elements, and engine wear and corrosion products may also contribute [1, 2].

Now commonplace for diesel applications, there has recently been increasing interest in applying the same types of filters to reduce PM emissions from gasoline engines as well [3]. In particular, the trend towards increased use of gasoline direct injection technologies, combined with the current and proposed implementation of strict particle mass and number emissions standards, have motivated serious interest in the use of gasoline particulate filters (GPF) as a viable means of reducing gasoline particle emissions.

Although fundamentally DPFs, and the currently proposed GPF technologies share many similarities, including the cellular ceramic wall-flow design, there are noteworthy differences in their operating environments and filter design parameters. Relative to the diesel operating environment, gasoline applications are generally characterized by hotter exhaust temperatures, stoichiometric (rather than lean) combustion, lower PM mass but higher PM number emissions, and smaller particles. Given the differences in the operating environment and exhaust characteristics, variations in the filter design parameters such as porosity, mean pore size, pore size distribution, and cell geometry between diesel particulate filters and their gasoline counterparts can be expected. However, similar to DPFs, particles accumulated on the GPF will also need to be oxidized, with the remaining incombustible material accumulating in the filter as ash.

Recently much progress has been made in understanding the impact of exhaust conditions and filter operating parameters, including regeneration strategy, on the accumulation and distribution of ash in diesel particulate filters. Given the fundamental nature of the work, it is expected that many of the same mechanisms may be equally applicable to gasoline particle filters as well. This study presents additional fundamental measurements, supported by theoretical analysis, which describes the primary mechanisms controlling the formation and subsequent transport or mobility of ash deposits in the particulate filter.

Effect of Ash Distribution on Filter Performance

Several studies have reported on the effects of ash distribution on DPF performance, specifically the impact of the ash on filter pressure drop. These studies have observed varying degrees of sensitivity to differences in the overall bulk distribution of the ash, whether accumulated in a layer on the channel walls or in a plug at the back of the channel, as well as the packing characteristics and nature of the ash deposits. The evidence to date points to a strong influence of the amount of PM accumulated in the DPF prior to full- or partial-oxidation, the details of the regeneration process, and the exhaust flow history of the filter, as key parameters controlling the ultimate distribution of the accumulated ash. However, the mechanisms governing the mobility of the ash from the channel walls to the plugs at the back of the filter have been, as of yet, unclear.

Differences in DPF pressure drop by more than a factor of two, for filters with nominally the same ash mass but different ash distributions, have been reported with DPFs obtained from vehicles operating in the field. Void regions in the ash plugs, as well as ash bridging were reportedly responsible for some of the more significant differences, attributed to incomplete regenerations over the lifetime of the filter [4]. Similarly, measurements from filters loaded with ash in the laboratory under controlled conditions showed an increase in pressure drop for DPFs exhibiting thicker wall ash layers and less ash packed in the channel end plugs [5]. Despite the increase in

filter pressure drop with increasing ash build-up, an increase in filtration efficiency has also been reported, due to the formation of a thin ash membrane on the channel walls [6].

Aside from experimental studies, simulations have been employed to quantify the impact of ash distribution on DPF pressure drop. Work by Gaiser et al. showed a reduction in filter pressure drop when all of the ash was accumulated in the back of the channel, relative to the same amount of ash distributed as a layer along the channel walls (considering the impact of the ash alone). On the other hand, when PM loading was also considered in the model, the opposite effect was observed [7]. More recent work by the authors employing a similar modeling approach showed the extent of the flow restriction, due to variations in ash distribution, to be quite sensitive to the specific ash properties [8].

Further, most of the theoretical models to date, utilize an assumed ash distribution profile, but do not account for transport of the particles along the DPF channels following initial deposition and agglomeration. This has been due, in large part, to a lack of information and understanding of the mechanisms responsible for the mobility or migration of ash in the filter. An initial framework was proposed by Konstandopoulous et al. which described ash removal from the channel wall in terms of a critical shear stress imposed by the flow, which, if exceeded, would result in particle detachment and subsequent transport [9]. A similar approach was applied in the development of a one dimensional (1-D) model in this study, with good agreement to experimentally measured DPF ash profiles. However, as the results of the experimental work will show, additional mechanisms also need to be considered.

Ash Formation and Transport

Considerable work has been done outside the engine and aftertreatment communities to explore powder flows and better understand nano-particle detachment and transport from surfaces. The findings of several fundamental particle transport studies were summarized and related to ash transport in diesel particulate filters in a recent paper by Dittler [4]. Further review of the powder transport literature has highlighted additional parameters which may be relevant to understanding ash transport in particulate filters.

Figure 1 presents a simplified free-body diagram showing the forces acting on a generic particle at rest on a surface, but exposed to an external cross flow [10]. In this case the primary forces acting on the particle include a horizontal component of the fluid drag force, F_D , a vertical lift force F_L , gravitational force mg (generally negligible), and an adhesive force F_A . In the case of a particle resting on a porous media through which the fluid flows (such as in the case of a particulate filter), the lift force, F_L , may be in the negative direction and correspond to the drag force imposed by the wall flow.

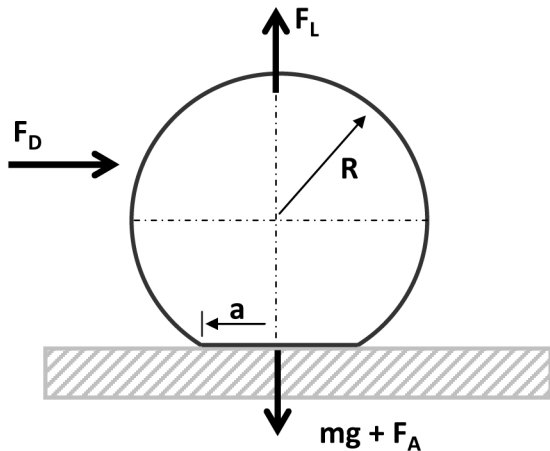


Figure 1. Forces acting on particle accumulated on filter surface. Schematic adapted from [10].

While the drag and/or lift forces can be estimated with a reasonable degree of certainty, given information on the flow field and particle size, defined in this case by its radius, R , quantifying the adhesive force is not nearly as straightforward, and is the focus of this investigation. For ash and soot deposited on a particulate filter, the adhesive force is thought to be dominated primarily by Van der Waals forces, although electrostatic and thermophoretic forces may play a role in some cases [11]. Furthermore, the magnitude of the adhesive force will be a direct function of the contact area, defined by the dimension, a , in Figure 1.

Ibrahim et. al. conducted a series of comprehensive fundamental particle transport studies utilizing spherical glass and stainless steel particles in the 30 - 72 μm size range, as well as a non-spherical Lycopodium spores in the 30 μm size range deposited on a glass slide. The particles were exposed to a range of flow conditions in order to investigate the various particle detachment and transport mechanisms, as well as the parameters influencing the adhesive force between the particles and glass surface. The work categorized the observed particle transport via three mechanisms: (i) particle lift-off, (ii) particle rolling, and (iii) particle sliding. Further, particle detachment increased with increasing free stream velocity and particle size [10].

Additional follow-on studies investigated the impact of humidity and residence time on the particle-to-surface adhesion forces using the same glass, steel, and Lycopodium spore particles. Humidity, was observed to play a significant role, above 30%, as was the particle residence time, particularly under humid conditions. In both cases, increasing humidity and increasing particle residence time on the surface resulted in an increased force of adhesion. Other parameters investigated included the influence of Reynolds number, whether laminar or turbulent, transient burst, and surface roughness [12, 13].

Of all the parameters investigated, the adhesive force between the particle and surface was found to be particularly sensitive to surface roughness. Relative to a nominally smooth surface,

increasing the variation in asperity heights on a small scale, the extent of which was partly dependent on the material, showed a large reduction in the force required to remove the particles [14]. Theoretical models, based upon the initial work of Greenwood and Tripp to describe the elastic contact of two rough surfaces have also been successfully applied to estimate surface roughness effects on particle detachment from surfaces [14].

Nitschke and Schmidt reported the development and validation of a particle transport model [15] against the experimental data provided by Ibrahim [10]. The model achieved high fidelity and accounted for surface roughness, fluid forces, and adhesive forces, taking into consideration Van der Waals forces and condensation under humid conditions [15]. Although much progress has been made in advancing the fundamental understanding of particle transport on flat, non-porous surfaces, additional complexity is introduced when soot and ash particles in the DPF are considered, along with the somewhat more complex flows around the particles and through the porous media.

While this study examined the macroscopic phenomena through a combined approach of in-situ optical measurements with actual DPF samples, and theoretical analysis of the near-surface flows, additional work by the authors has focused on directly quantifying the inter-particle adhesion forces between soot, ash, and the filter media using atomic force microscopy [11]. The results provide an enhanced conceptual understanding of the physical mechanisms controlling ash and soot transport in particle filters, which should be equally applicable to describe the processes occurring in both diesel and gasoline applications.

EXPERIMENTAL PROCEDURES

This work utilized particulate filter core samples mounted in a fixture, with optical access to a small portion of a single filter channel. In this manner, the soot oxidation, ash formation, and transport processes could be observed with the aid of a stereomicroscope, in situ. All experiments were conducted with a catalyst bench reactor to enable careful control and manipulated of the regeneration and exhaust flow conditions. The experimental setup and procedures are similar to those described in a previous study by the authors [16], and are briefly summarized here.

Filter Samples and Loading Conditions

The filter core samples used in this study were cut from full-size D5.66" (14.38 cm) $\times$ L6" (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 to facilitate soot oxidation.

The cores for the bench reactor and flow tests measured approximately 0.875" in diameter (2.22 cm) and spanned the entire length of a 6" (15.24 cm) DPF. The small cores were wrapped, using a commercial intumescent mat, and canned in stainless steel sample holders. The specially-designed sample holder was used to facilitate installation of the core samples on the flow bench for pressure drop measurements and performance characterization, while at the same time enabling optical access to a portion of the exposed filter channel.

Soot loading of the filter samples was accomplished by diverting a small portion of exhaust from a 2009 Yanmar L100 to flow through the sample fixture. 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, the ash content of the soot was increased to allow for visualization of the ash formation and transport processes in the filter using a burner-based accelerated ash loading system in conjunction with the diesel engine.

Details of the accelerated ash loading system used in this work can be found in several previous publications [17, 18, 19]. In summary, the system allows for control and acceleration of certain oil consumption mechanisms, to simulate engine-out ash and PM emissions as closely as possible. Previous studies showed similar exhaust ash emissions characteristics between the accelerated ash loading system and a Cummins ISB diesel engine [19]. Additionally, 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 sets of filters [17].

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 detailed composition of which can be found in [20]. Conventional ultra-low sulfur diesel (ULSD) was used in both the engine and burner system for all of the tests.

Optical Sample Fixture and Test Conditions

Figure 2 shows the optically-accessible filter core sample fixture, which enables optical access to a small portion of the outermost filter channel. Figure 2(b) shows how 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 regeneration and particle transport processes on the channel surface. An electrical heater (not shown) is used to minimize the heat losses near the viewing window. The fixture provides a rectangular field of view approximately equal to the width of the channel (1 mm) and several millimeters long.

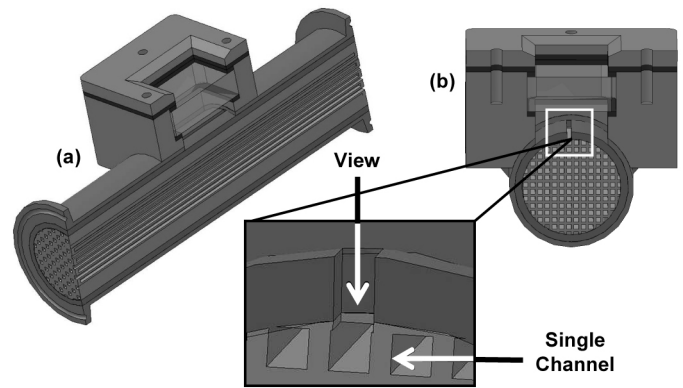


Figure 2. DPF core sample fixture with optical access (a) and detail showing field of view into single channel (b).

The filter core sample with optical access also allows for soot loading on the engine, and regeneration and flow studies on the catalyst bench reactor, shown in Figure 3. This system enables control of the inlet gas composition, flow and temperature, as well as measurements of filter pressure drop and temperatures. The exhaust gas composition upstream and downstream of the core sample was monitored using an MKS FTIR analyzer.

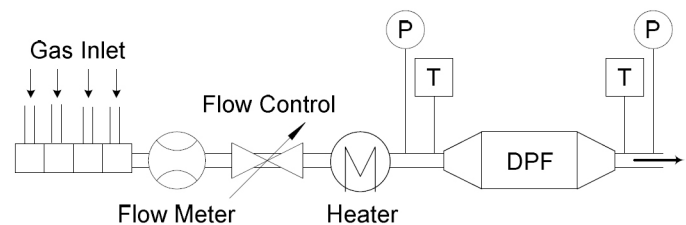


Figure 3. Flow bench and reactor setup for controlled regeneration and ash transport studies with optically accessible DPF samples.

Unless otherwise specified in the discussion of the results, the filter regeneration was carried out following soot loading with engine-out exhaust to a level of approximately 4 g/L. The gas composition used with the bench reactor during regeneration consisted of 500 ppm NO with dry air balance, as the objective of this work was to visualize the fundamental physical particle transport processes occurring during the regeneration event, as opposed to characterizing the catalyst performance.

Following complete or partial regeneration of the filter core sample, the system was cooled to room temperature, prior to initiating a controlled, step-wise increase in flow. The test procedure is schematically depicted in Figure 4. Dry, filtered shop air was supplied to the flow bench, and the flow rate was gradually increased from a starting value of approximately 20,000 hr⁻¹ to a final value in excess of 160,000 hr⁻¹. While the upper end of the flow range may be somewhat unrealistic for on-vehicle conditions with a standard size DPF, it does provide data useful to quantify the impact of elevated flow on particle mobility and transport in the filter channels.

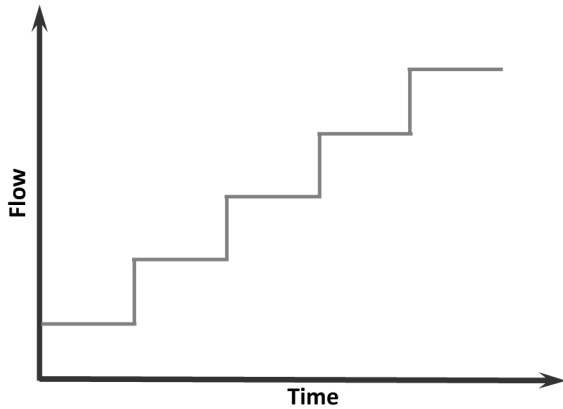


Figure 4. Step-wise increase in flow through optical DPF samples following full- or partial-regeneration.

The typical step sizes between flow set points in Figure 4 ranged from $10,000 \text{ hr}^{-1}$ to $20,000 \text{ hr}^{-1}$. A constant flow was maintained at each step for approximately 30 seconds prior to systematically increasing the flow rate, and the frame capture rate with the stereo microscope was one frame per second.

THEORETICAL FRAMEWORK

Two approaches were employed to develop a theoretical framework to interpret the experimental observations, and provide additional analysis of the key parameters which could not be directly measured in the experiments. An initial one-dimensional (1-D) model was developed, based on a simple force balance on the soot/ash particle and an empirical parameter to describe the critical particle removal/detachment stress. Given the importance of the flow fields in the vicinity of the particles on the filter surface, a more detailed three-dimensional (3-D) computational fluid dynamics (CFD) model was employed to estimate the drag and lift forces imposed on the particle by the exhaust flow.

One-Dimensional Model

A one-dimensional flow model was formulated to describe the flow inside the DPF channels for a given ash cake layer profile and end-plug length, in order to estimate the shear stress imposed by the flow on the surface particles. The governing equations for the classical 1-D models developed by Konstandopoulos and Johnson and [21, 22, 23] were modified to also include variations in the deposit cake layer thickness and distribution profile. The governing equations can be reduced to one non-linear ordinary differential equation (ODE), which is solved using a pseudo-time loop method by adding a time derivative term in the equation. The details of this flow model, and specifics related to the derivation, can be found in a previous publication by the authors [8]. Figure 5 presents the ash deposit geometry considered in the model and the relevant flow parameters.

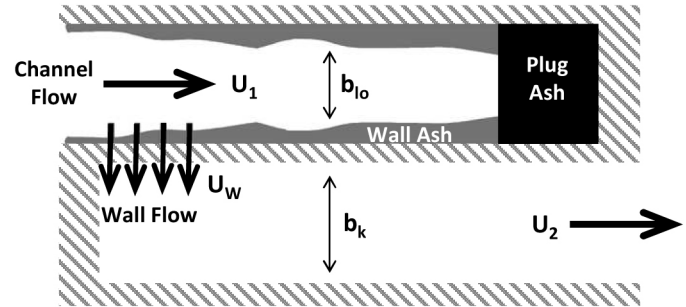


Figure 5. Illustration of geometry and parameters considered in one-dimensional filter particle transport model.

As the 1-D model represented an initial approach to estimating the flow-induced shear required to induce particle transport from the channel walls to the back of the channel, the following simplifying assumptions were made:

1. Ash particles transport was only considered following complete soot oxidation, and the transport of soot along with the ash was not included in this initial model.
2. The local shear stress was estimated from the local mean channel flow velocity, which was calculated from the 1-D flow model.
3. Once the local flow shear stress exceeds the particle's critical detachment stress, the particle begins to move.
4. Particle re-deposition was accounted for by assuming the particle will re-deposit in a position where the local flow shear stress equals the particle's critical re-deposition stress.

The detachment and re-deposition stresses for the particles were calculated from experimentally measured ash distribution profiles, to fit the measurement data. The local flow shear stress was calculated from the 1-D model as follows:

$$\tau = \frac{F\eta u}{4b_{lo}}$$

Equation 1

In Equation 1, F is the laminar channel flow friction factor, η is the gas viscosity, u is the inlet channel local mean axial velocity which is a function of axial position, and b_{lo} is the open width of the inlet channel considering the ash deposit profile.

The basic structure of the 1-D ash distribution model is described as follows. First, soot deposition is simulated based on the 1-D flow model. Second, a regeneration model considers the increase in ash layer thickness after each regeneration cycle. The ash transport/mobility is calculated through a trial-and-error process. At each cycle, the flow field and shear stress is calculated and compared with the particle detachment criteria for each local position along the length of the channel. If the particle moves, its new re-deposition location will be the position where the flow shear stress equals the particle's critical re-deposition stress. This cycle is repeated until the particles cease to move, and an equilibrium layer