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Near-wall dispersion, deposition and transformation of particles in automotive exhaust gas aftertreatment systems

Henrik Ström[a,*], Jonas Sjöblom[b], Ananda Subramani Kannan[a], Houman Ojagh[c], Oskar Sundborg[b], Jan Koegler[d]

[a] Division of Fluid Dynamics, Department of Mechanics and Maritime Sciences, Chalmers University of Technology, SE-412 96 Gothenburg, Sweden
[b] Division of Combustion, Department of Mechanics and Maritime Sciences, Chalmers University of Technology, SE-412 96 Gothenburg, Sweden
[c] Division of Chemical Engineering, Department of Chemistry and Chemical Engineering, Chalmers University of Technology, SE-412 96 Gothenburg, Sweden
[d] Volvo Group Trucks Technology, Powertrain Advanced Engineering, Dept. BF61500 Exhaust Aftertreatment, Sven Hultins gata 9B, SE-412 88 Gothenburg, Sweden

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ABSTRACT

Combustion-generated nanoparticles are present in exhaust gas aftertreatment systems in the approximate size range 1–1000 nm. Successful optimization of aftertreatment systems for pollution control relies on the existence of numerical tools to predict the momentum, heat and mass transfer between these types of particles and the surrounding gas phase. Such tools can only be readily obtained if our fundamental understanding of the phenomena pertaining to the particle behavior is correct.

The small nano-particulates in automotive exhaust gas aftertreatment systems are typically described as spherical and inert, closely following the gas phase streamlines apart from a superimposed Brownian motion. However, for real particulate matter, produced by an internal combustion engine, we show that the deposition in an automotive catalyst substrate cannot in general be well described by the aforementioned modeling approach, as particle transformations become active inside the substrate channels, altering the apparent deposition efficiency. A conceptual model is proposed that is able to explain the initially observed discrepancies between measurements and simulations, by describing the particulate matter as a mixture of three different types of particles: truly inert particles, semi-volatile particles and completely volatile particles. The conceptual model is corroborated by experimental and numerical investigations into the behavior of truly inert particles in automotive catalyst substrates.

Finally, the possibility to use the model for in-situ characterization of particulate matter is demonstrated. For the first time, data is presented to support the hypothesis that differences in particle properties, as characterized in this way, have a meaningful correlation to particle reactivity in e.g. oxidation experiments. In other words, particle reactivity may be assessed indirectly by investigations of particle mobility. It is also shown how the pressure drop through a bed of deposited nanoparticles will differ depending on the properties of the deposited particles, as characterized by the conceptual model.

1. Introduction

Exhaust gas aftertreatment systems for typical vehicle applications contain large numbers of very small particles generated in the cylinders of the internal combustion engine (Kittelson, 1998). These particles are to be removed in some way, for example via collection in a particulate filter, prior to their release into the ambient. One major drawback with employing such filters in exhaust gas pipes is the incurred pressure drop, which translates into a fuel penalty. Optimized filter design is therefore an important topic within research on automotive particulate emission abatement (van Setten et al., 2001). Other technological challenges with small, engine-generated particles in general aftertreatment systems include fouling of heat transfer surfaces due to Brownian and thermophoretic deposition (Abd-Elhady et al., 2010), possible catalyst deactivation by deposition (Larsson, 2007), and clogging of porous materials – in particular catastrophic clogging of filters due to the melting of deposited ash during regeneration (Liati and Dimopoulos Eggenschwiler, 2010; Wurzenberger and Kutschi, 2007). To arrive at more advanced and suitable solutions to these challenges, a better understanding of how these small particles are transported through the aftertreatment system is needed.

The particulate matter generated during combustion is typically

* Corresponding author.
E-mail address: henrik.strom@chalmers.se (H. Ström).

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extremely small, ranging from a few nanometers to hundreds of nanometers, and are present in high number concentrations. The particle size distribution is typically bimodal, and it is therefore customary to classify the nanoparticles as belonging to either the nuclei mode (5–50 nm, consisting of either volatile organic compounds that form during cooling, or solid carbon and metal compounds) or the accumulation mode (100–300 nm, consisting of larger agglomerates formed from nuclei mode particles) (Kittelson, 1998). If accumulation mode particles deposit on surfaces and are later re-entrained, a trimodal distribution may appear where the third group represents the so-called coarse mode (1–10 μm) (Kittelson, 1998).

Visualizing the motion of individual nanoparticles in exhaust gas aftertreatment systems is difficult experimentally due to their small size and the lack of optical access. A straightforward way to characterize their motion in and with the gas phase is however to quantify the extent of particle deposition in a given system (Johnson and Kittelson, 1996; Sjöblom and Ström, 2013). This can be accomplished by inferring the number of particles deposited within a system by relating the number of particles entering the system to the number exiting. In this way, information about the particle motion is obtained indirectly. Mathematical models developed for detailed descriptions of particle motion can then be validated from the predicted deposition efficiencies in various catalyst substrates.

The physics controlling the nanoparticle motion is strongly dependent on the exhaust gas flow regime. Wherever the flow is turbulent, the concentration profile of small particles tends to be quite even throughout the bulk, with steep gradients in the near-wall regions (Soldati and Marchioli, 2009; Li and Ahmadi, 1992). As the particles are of nanometer size, and the walls are never perfectly smooth at these spatial scales, surface forces are large and the particle-wall adhesion is strong. In addition, in the porous walls of a filter or catalyst substrate, the aerodynamic forces that could possibly cause particle re-entrainment are relatively small. Particles therefore typically stick to the wall once they deposit. For particles large enough to possess appreciable inertia, there is a trend to near-wall accumulation due to the wall-normal gradients in the turbulent fluctuating velocities (Soldati and Marchioli, 2009; McLaughlin, 1989). Particles accumulated close to the wall in such processes will either eventually be brought back into the bulk by an ejection sweep or have time to deposit due to Brownian motion (Brooke et al., 1994). Deposition of nanoparticles in turbulent flow has been studied extensively in the literature (Brooke et al., 1992; McLaughlin, 1994; Ounis et al., 1991a, 1991b; Chen and McLaughlin, 1995; Kallio and Reeks, 1989; Wang and Squires, 1996a,b).

In the sections of the exhaust gas aftertreatment system where the flow is laminar, i.e. mainly inside the channels of the catalyst and filter substrates, the boundary layers are thicker and the radial transport of particles is much smaller. This transport can be significantly enhanced by provoking a secondary flow in the cross-section normal to the main gas flow direction (Ström et al., 2010). The most successful measure to create suitable secondary flow patterns is thus to introduce obstacles (bumps and/or protrusions) in the channels, to have them deviate from straight ducts (Sjöblom et al., 2017a). Such substrate designs are sometimes dubbed “turbulent” even though the mixing is not caused by actual turbulence (Ström et al., 2012).

Computational optimization of substrate and channel designs intended to either maximize or minimize the deposition of nanoparticles onto the walls is relatively straightforward if the particles are treated as inert spheres. However, this is a major assumption that is in direct conflict with the current phenomenological understanding of how realistic engine-generated particulate matter is constituted: spherules that come together to form larger fractal-like agglomerates with adsorbed or condensed hydrocarbons (HCs) over the surfaces (Kittelson, 1998; Sjöblom and Ström, 2013; Twigg and Phillips, 2009). If these hydrocarbons desorb, it is not unthinkable that the particle mobility will be affected. Such desorption could for example be triggered by adsorption of hydrocarbons from the bulk to the walls of the catalyst substrate, which would lower the background HC concentration and thus start to drive off HCs from the particulates. When it comes to droplets in automotive exhaust gas aftertreatment systems, it is well known that they change size, and therefore also their diffusivity, as they evaporate or react (Lundström et al., 2013). There is no reason not to expect that similar effects could be important for nanoparticles as well.

The main purpose of the current paper is thus to question the applicability of the inert particle assumption for realistic engine-generated particles, to explore better models for describing the size changes undertaken by these nanoparticles, and to highlight the new routes to understanding realistic nanoparticle motion opened up by these aspirations. More specifically, we will here illustrate for the first time how a conceptual model for particle transformations can be used not only to more accurately predict nanoparticle motion, but also for in-situ characterization of particulate matter with regard to reactivity (in subsequent oxidation experiments) and aerodynamic properties (during packing to a bed of deposits in a particulate filter).

2. Materials and methods

2.1. Experimental

The present work is mainly concerned with the various strategies to mathematically and numerically describe the motion, deposition and transformations of particulate matter in exhaust gas aftertreatment systems. Hence, the experimental overview will be rather brief and the interested reader is directed to our previous publications for additional details (Sjöblom and Ström, 2013; Sjöblom et al., 2014a, 2014b, 2017a; Sjöblom, 2013).

The experimental setup used for the acquisition of most of the data in this work is an Exhaust gas AfterTreatment System (EATS) rig (Sjöblom and Ström, 2013; Sjöblom, 2013). In this setup, a portion of the exhaust from a diesel engine run at constant load is fed through a heater and a mass-flow controlled air-addition setup into an open ceramic catalyst substrate. The particle size distributions before and after the substrate are measured by a fast particle analyzer (DMS500 from Cambustion) to enable determination of the size-resolved particle deposition efficiencies. To obtain a good signal-to-noise ratio, the retention time is long enough (i.e. longer than in actual on-road conditions) to achieve a considerable particle deposition.

The EATS was later expanded with the possibility to disperse sodium chloride nanoparticles by atomization of an aqueous NaCl solution with a Topas ATM230 atomizer (Sjöblom et al., 2014b). A full-scale setup was also developed for evaluation of a novel metallic catalyst substrate based on short retention times and high linear velocities through the channels (Sjöblom et al., 2017a).

In this work, we present new experimental data obtained in a different rig where the EATS methodology has been applied, as shown in Fig. 1 (Sjöblom et al., 2017b). The exhaust from a heavy-duty diesel engine is passed through a full-scale diesel particulate filter (DPF) that is connected in parallel with a cartridge of lab-scale DPFs. The DPF cartridge can either be alone in its pipe section, or have a diesel oxidation catalyst (DOC) upstream. The purpose of this setup is to enable longer filter loading experiments wherein real engine-generated particulate matter is loaded into a lab-scale DPF at the same driving pressure difference as a full-scale DPF. The upstream DOC is included to allow characterization of the particulate matter, as shall become clear in Section 3.3. Finally, oxidation of the particulate matter in the lab-scale DPF can be monitored to secure information on the reactivity of the particles collected in different types of filters (e.g. different catalytic coating).

The loading of the lab-scale DPFs with particles was done through a set of different driving cycles including steady state and transient operation. Each set of cycles lasted 35 min and was repeated eight times. The pressure over the full-scale DPF varied between 0–9 kPa and constituted the driving force for soot loading in the small mini-DPFs.