

Modeling the Emissions Control Performance of a Catalyzed Diesel Particulate Filter (CDPF) System for Light Duty Diesel Applications

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ABSTRACT

The use of catalyzed diesel particulate filter (CDPF) systems in light duty diesel (LDD) vehicles is becoming increasingly common. The primary functions of the system are to remove carbon monoxide (CO) and hydrocarbons (HC) from the vehicle exhaust stream, while simultaneously reducing the level of particulate matter (PM) emissions to ambient background levels. These systems can comprise either a separate diesel oxidation catalyst (DOC) and a downstream CDPF, or a single unit CDPF with the DOC functions incorporated within the CDPF. The single CDPF unit provides higher regeneration efficiency as it is located nearer to the engine and also cost benefits, as only a single unit is required compared to the alternative separate DOC and CDPF arrangement.

A model describing the performance of the single unit CDPF for emissions control has been developed, with particular emphasis on achieving predictions of the CO and HC emissions over transient vehicle drive cycles. The model comprises a 1-D DPF model, with a description of soot accumulation and removal, and CO, HC and NO oxidation over the catalyst coated on the filter wall. Langmuir-Hinshelwood expressions were employed to describe the CDPF reaction kinetics.

The model has been validated using transient European (ECE/EUDC) drive cycle data. The model has been developed to incorporate diffusion of the exhaust

components to the catalyst surface, akin to mass transfer in a monolith substrate. This enabled a study of the effect of precious metal catalyst loading and precious metal catalyst distribution within the filter on the CDPF emissions performance. The model can be used as a tool to aid optimized CDPF catalyst design as well as sizing and positioning of a CDPF on different vehicle applications.

1. INTRODUCTION

Emission control has been increasingly integrated into vehicle design for the past three decades. For example, gasoline passenger vehicles are fitted with monolith type three-way catalysts to reduce the emissions of carbon monoxide (CO), hydrocarbons (HC) and nitrogen oxides (NOx). More recently, to meet tighter diesel emissions legislation worldwide, the trend of incorporating a catalyst has extended to diesel vehicles. Diesel catalysts reduce CO and total HC emissions, while reduction of NOx and particulate matter (PM) has been mainly achieved by advanced engine designs.

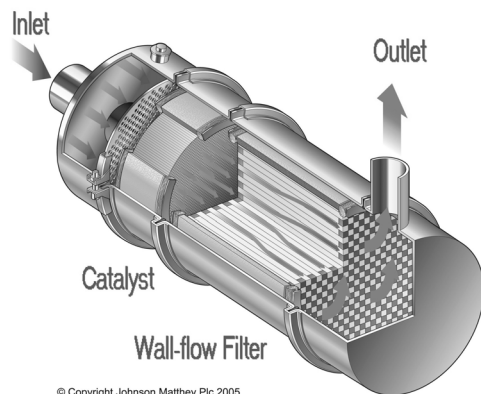
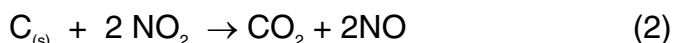
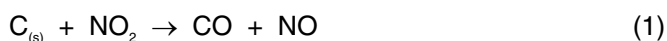


Figure 1. Schematic representation of the CR-DPF system.

To meet future diesel emissions legislation, diesel particulate wall-flow filters (DPF) can be used and in the heavy duty diesel (HDD) sector, the continuously regenerating diesel particulate filter system (CR-DPF or CRT[®]) shown in Figure 1 [1] has been employed extremely successfully. Here an upstream diesel oxidation catalyst (DOC) removes CO and HC from the exhaust stream, while it also converts some of the engine out NO to NO₂, for subsequent reaction with PM in the downstream filter (reactions 1 and 2). In many applications regeneration occurs by a continuous process, and the system needs no servicing or complex active regeneration strategy [2].



Within the light duty diesel (LDD) sector, four main types of filter systems have been developed as shown in Figure 2. In the first system (Figure 2i), like the HDD CR-DPF system, a diesel oxidation catalyst is positioned upstream of a DPF and use can be made of NO₂ to passively remove PM from the filter.

However, in many situations on light duty vehicles, the NO_x/PM ratio is not sufficient to remove all the collected PM or the operating temperature is not high enough for the reaction between NO₂ and PM to occur at a fast enough rate, and therefore a totally passive application of the CR-DPF is not possible. In these more challenging circumstances an active regeneration strategy is applied to the system whereby the engine will periodically raise the temperature of the exhaust gas to that required to burn the PM more rapidly. In order to further improve both the passive and active regeneration of the filter system a catalyst coating is applied to the filter (Figure 2ii). Improved performance is achieved by re-oxidation of NO within the catalyzed filter to form further NO₂ that subsequently reacts with more PM, and

by direct catalysis of the oxygen-PM reaction. The resulting DOC + CDPF system has a wider operating window than is the case for the DOC + DPF system [3].

The system shown in Figure 2iii does not require the separate upstream DOC but here the DOC is replaced by incorporating the DOC coating on the filter itself [4-10]; this system is known as a single-unit catalyzed diesel particulate filter (SCDPF). One benefit of this system is that because CO, HC and PM reduction are all achieved on a single component, rather than two as is the case for the DOC + CDPF, there are potential packaging and cost benefits. Another benefit results from being able to locate the single unit closer to the engine compared to filter systems that use upstream DOC components: in this closer location there is less heat loss in the pipework connecting the filter to the engine and this results in more efficient regeneration of the filter.

A further system is shown in Figure 2iv, where a small DOC is positioned in front of the CDPF. The main advantage of this system is that the DOC has a much lower thermal mass than the CDPF, and therefore heats up much more quickly than the CDPF. This results in improved light-off during warm-up of the system. However, the DOC must be of small volume to still allow the CDPF to be mounted close to the engine and thus minimize heat losses, whilst still being of sufficient volume to provide good HC and CO light-off performance. The small DOC is not able to provide the high levels of CO and HC oxidation required especially under higher flow rate conditions. Under these conditions, the downstream CDPF, that also incorporates a DOC coating, can provide very high conversion of CO and HC. A feature of this system is that it is a combination of both the small volume DOC and the specialized coating on the CDPF that provides the overall high CO and HC conversions required.

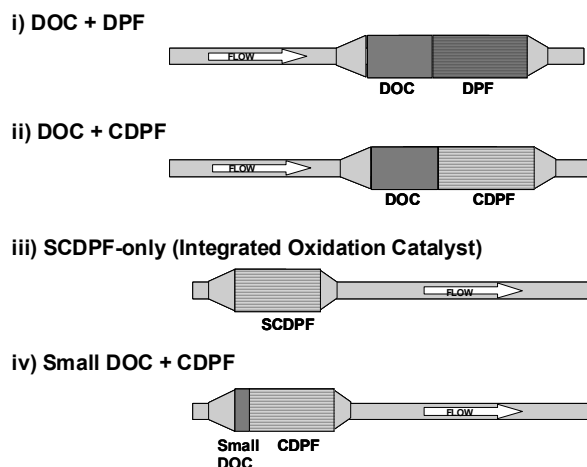


Figure 2. Diagram showing different diesel particulate filter systems.

Clearly, to design a CDPF system for use in LDD applications, a number of factors need to be considered. Physical properties such as filter size, substrate type (e.g. cell density, wall thickness, material), and positioning relative to the engine are all important aspects of system design. Within the CDPF the catalyzed reactions with the gaseous exhaust components such as CO, HC and NO need to be considered as well as the effect of accumulation and removal of PM, and the effect this process has on the gaseous emissions. For example, it is important to optimize the reactions shown in Equations 1 and 2 in order to maximize the amount of passive cleaning of the filter system, and to minimize the number of active regenerations required.

The use of computer simulations to study the performance of different CDPF designs is very beneficial in order to assess a wide range of scenarios quickly and cost effectively. This paper describes the development of a CDPF model capable of simulating emissions control performance, starting with a model incorporating only convective transport processes. This model was effective for simple catalyzed systems, but was inadequate with more complex zone catalyzed filters. Therefore a second-generation model is also described, wherein the reactants reach the catalyst by diffusion; this model is capable of predicting the emissions performance of zoned systems. The model includes a detailed description of the chemical kinetics of the salient reactions. The simulated emissions from the model have been compared to engine data, and a good agreement was obtained. The model can be used to aid component designers optimize the CDPF emissions performance for a wide range of LDD vehicles, while also accounting for packaging and cost constraints.

2. EXPERIMENTAL

2.1. ENGINE BENCH EXPERIMENTS

Experiments were carried out using catalysts coated onto standard silicon carbide (SiC) diesel particulate filter (DPF) substrates. The CDPF systems tested were all aged for 100,000 km.

Engine bench light-off experiments were carried out operating at a constant engine speed. The exhaust temperature was increased by increasing the engine load. European drive cycle (ECE/EUDC) tests were carried out on a vehicle driven on a test bed according to standard procedures. Diesel fuel with sulfur level < 50ppm was used during these tests. The fuel was purchased from a standard refinery source.

Gaseous emissions were measured raw before and after the catalyst system, using a range of analyzers:

chemiluminescence for NO and NO₂, heated FID (Flame Ionization Detector) for HC, NDIR (Non-Dispersive Infra Red) analyzers for CO, CO₂ and N₂O, and a paramagnetic detector for O₂.

3. RESULTS AND DISCUSSION

3.1. CDPF MODEL DEVELOPMENT

Wall flow filter models are widely available in the literature [11-20], and are mainly based on the work of Bissett [11]. More recently, these models have been modified and supplemented to describe catalyzed wall flow filters [9,10]. However, the DPF modeling to date has tended to concentrate on backpressure and regeneration processes. Emission control modeling has received some attention, but until recently this had focused mainly on monolith-type catalyst systems [20-23]. However, recently Koltsakis et al. [24] used modeling to optimize emissions reduction over a zoned catalyzed DPF, looking at the effect of zoning. Also, Ahmadinejad et al. [25] published a brief description of the modeling of zoned CDPF systems. These studies aside, there has been little activity on modeling the performance of catalyzed soot filters as emissions control devices.

The DPF model used in this work is based on that described in [11]; this model solves the mass, momentum and heat balances, and calculates pressure drop across the filter wall and soot cake. A schematic diagram of this section of the model is shown in Figure 3. The relevant equations can be found in the publications already listed.

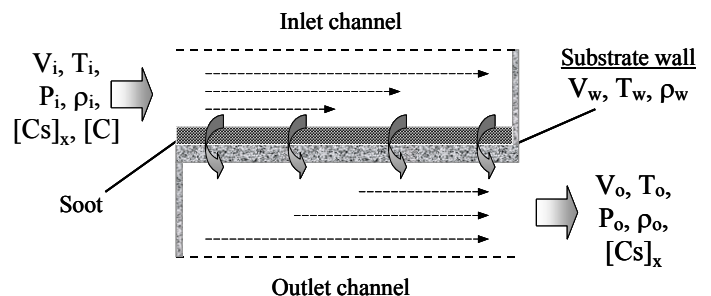


Figure 3. Schematic diagram of the DPF and the variables involved in the DPF model.

Some of the most important assumptions made in the DPF model used here are:

1. uniform flow distribution at the filter entrance.
2. no radiative heat losses.
3. inlet channel narrowing due to soot deposition on the wall is taken into account.

- soot is deposited on the filter wall in accord with the velocity profile (i.e. higher momentum of soot particles relative to the gas is not considered).
- negligible radial temperature and concentration gradients.
- inertial contributions to the pressure drop due to expansion/contraction at the filter exit/entrance are included.

The equations were discretized in the axial direction. A number of boundary conditions were included in the model, and ordinary differential equations (ODEs) constituting this boundary value problem (BVP) solved using a standard BVP solver.

The method used to model the processes and reactions occurring in the filter wall was similar to that described by Huynh et al. [9] and Konstandopoulos et al. [12]. These previous studies were interested in soot filtration and mass balance throughout the filter wall. Here this approach has been adopted and extended to a feed representative of a full diesel exhaust mixture (with the exception of sulfur species).

The wall is discretized both lengthwise, in the axial direction (x), and through the wall (y), as shown in Figure 4. In each element, (x,y), mass balances and rate equations for the diesel exhaust species, CO, C₃H₆, C₇H₈, n-C₁₀H₂₂, NO, NO₂, and O₂, are applied. Rate equations, for reactions 3-7, were of the Langmuir-Hinshelwood type employed in previous studies [20-23]. Diesel exhaust is a complex mixture of hydrocarbons; propene, toluene and n-decane were chosen to be representative of the hydrocarbons present in the diesel exhaust.

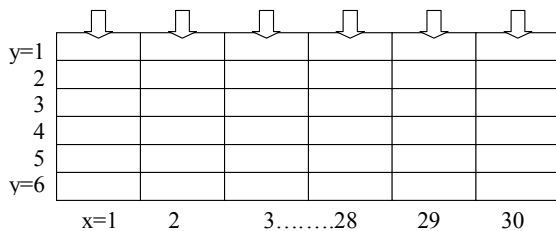
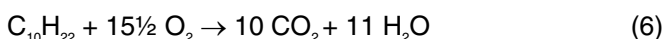
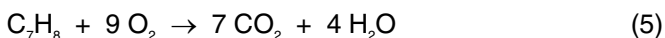
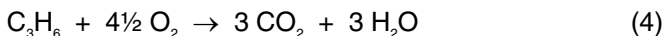


Figure 4. Schematic diagram of the CDPF, showing filter wall discretization in the x and y directions.



The Langmuir-Hinshelwood equations are of the basic form:

$$R_i = \frac{A' \exp\left(\frac{-E_a}{RT_w}\right) [A]^m [B]^n}{(1 + K_a [A])^p (1 + K_b [B])^q} \quad (8)$$

In the case of NO oxidation to NO₂ it is necessary to include a term to describe the thermodynamics of the reaction, and a modified equation was used (9):

$$r_{(\text{NO}-\text{O}_2)} = A'_{\text{NO}_2} \exp\left(\frac{-E_a}{RT_w}\right) \left(p_{\text{NO}} p_{\text{O}_2}^{0.5} - \frac{p_{\text{NO}_2} p_{\text{atm}}^{0.5}}{K_{\text{eq}}} \right) \quad (9)$$

In addition, PM oxidation rates are included in accord with equations 1 and 2, since these reactions impact the emissions of NO/NO₂ and CO/CO₂. Rate equations were of the form:

$$R_i = A' \exp\left(\frac{-E_a}{RT_w}\right) [\text{oxidant}] \quad (10)$$

This equation is integrated over the soot layer to account for the depletion of oxidant as the exhaust gas passes through the soot. This has the consequence that the total rate of soot oxidation depends on the thickness of the soot layer and becomes zero if the soot is completely removed.

In this way, the wall model takes the concentrations of species in the inlet channel, and calculates the concentrations throughout the filter wall, and those exiting the wall and entering the outlet channel.

3.2. MODEL VALIDATION USING ENGINE LIGHT-OFF DATA

Kinetic parameters were derived from detailed examination of engine light-off test data, and previous experience with similar catalytic systems applied to monolith substrates. Thus activation energies, pre-exponential factors and Langmuir-Hinshelwood constants for the different reactions 1-7 were obtained, with particular emphasis on reactions 3-7, since this study is mainly concerned with the performance of the CDPF as a gaseous emissions control device.

Figure 5 shows the results obtained for an engine light-off experiment performed on an engine with a 5.66" x 6" 200/12 SiC CDPF system.