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Application of a methodology to assess the performance of a full-scale diesel oxidation catalyst during cold and hot start NEDC drive cycles

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ABSTRACT

An experimental technique is demonstrated, in which the spatial variations of temperature inside a diesel oxidation catalyst (DOC) can be measured during an NEDC drive cycle. This leads to the development of a methodology which enables more scientific measurements to be performed on such a catalyst when it is connected to a live diesel engine. During hot and cold start NEDC drive cycles, measurements were performed on a Pt on alumina DOC connected to a Ford 2.0 litre diesel engine. Under these transient conditions, emission data was gathered (pre- and post-DOC), and this was also translated into cumulative emissions over the drive cycles. In addition, valuable data was gathered on the spatial temperature variations in the DOC during such a drive cycle and it is shown how, under certain conditions, the temperatures start to increase at the back-end of the catalyst and then move towards the front. This data, in combination with other measurements, helps to understand the performance of the DOC and can also be used to test the robustness of mathematical models of such systems.

Keywords: Diesel oxidation catalyst; NEDC drive cycle; Catalyst modelling; Diesel engine

1. Introduction

It is well known that catalysts are extensively used to control exhaust emissions from vehicles in a variety of different systems (e.g. Lox and Engler, 1999; Twigg et al., 2006). For example in the form of: three-way catalysts; diesel oxidation catalysts; selective catalytic reduction catalysts; NO_x storage catalysts; and in catalytic particulate filters. Being realistic when looking into the future, it is clear that a few more decades will pass before the use of carbon based fuels in internal combustion engines starts to be replaced at a more significant level by electric and hydrogen-powered vehicles. In the meantime, as developing nations continue to grow, the number of cars will increase in the world, and demand for the limited resources of precious metals used in catalytic systems to control emissions from vehicles will increase. Also, as government legislation to reduce emissions from vehicles becomes stricter, the car manufacturers will need to improve the design of such catalytic systems and how they are integrated with the engine management system in the vehicle.

Although the literature on the subject of catalytic systems to control vehicle emissions is vast, there is still plenty of scope to improve the design of such catalytic systems, as was evident from the 2nd International Symposium on Modelling of Exhaust – Gas After-treatment (MODEGAT II). In the preface to the special papers from the conference (Deutschmann et al., 2012), there was a clear recognition that:

- the design and optimisation of a catalytic converter is challenging (due to the complex interactions between chemical reactions and mass and heat transfer),

Abbreviations: NEDC, New European Drive Cycle; UDC, urban drive cycle; EUDC, extra urban drive cycle; CFD, Computational Fluid Dynamics; DOC, diesel oxidation catalyst; TWC, three way catalyst; CO, carbon monoxide; THC, total hydrocarbon; NO_x, oxides of nitrogen (NO and NO_2); EGR, exhaust gas recirculation; SCR, selective catalytic reduction; FTP, federal test procedure; GHSV, gas hourly space velocity.

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- experimental test bench measurements are needed (but they are expensive and time-consuming),
- when experiments are performed, they are difficult to interpret (because of the different chemical and physical processes taking place inside the catalytic structures).

In this paper, we consider the use of a catalyst to control the emissions of carbon monoxide (CO) and total hydrocarbons (THCs) from a diesel engine, using a device known as a diesel oxidation catalyst (DOC), and advance knowledge in the field of experimental measurement to support the challenges listed above in Deutschmann et al. (2012). The DOC consists of a catalyst support structure through which the exhaust gas flows. For example, a monolith structure is often used, which consists of a large number of parallel channels that are coated with a washcoat (e.g. γ-alumina) providing the high surface area that in turn supports the active catalytic ingredient(s), e.g. Pt, a system studied in this paper.

The reactions that take place are complex, but the main oxidation reactions may be simplified by the following overall expressions:

$$2CO + O_2 \rightarrow 2CO_2 \quad (1)$$

$$C_xH_y + (x + (y/4))O_2 \rightarrow xCO_2 + (y/2)H_2O \quad (2)$$

In the DOC, under oxidising conditions, some of the NO will be converted to NO_2, which could then be important in any selective catalytic reduction (SCR) device that may follow to reduce NO_x emissions. On a diesel engine, provided the electronic engine management system is functioning properly and the engine is warm (operating temperatures are adequate), a DOC is capable of reducing CO, and THCs emissions by about 99%. However, on engine start-up (when cold), they do not work effectively until the catalyst 'light-off' temperature is achieved, and therefore a significant amount of pollutants can be emitted during this heat-up period (e.g. Koltsakis and Stamatelos, 1997; Lewis et al., 2011). According to data presented in Carberry et al. (2005), about 70% of the THCs emissions are produced during this period on the New European Driving Cycle (NEDC) for a diesel engine. This is known as the cold-start effect, and is discussed in detail in many publications, e.g. Ashley (1996), Iliyas et al. (2007) and Nagashima et al. (2004).

Although DOCs are already used for the control of CO and hydrocarbon emissions from vehicles, there is still plenty of scope to improve their design. At present, catalyst development usually starts with bench-top laboratory scale experiments using powdered catalysts, and/or small sections of coated monolith. These samples are then mounted in small tubes (the reactor), and experiments are then performed using a synthetic gas mixture to represent the exhaust gas. Next, the information gathered from such experiments, often in combination with mathematical modelling techniques, leads to the design of a DOC, which is then made and tested on a real exhaust system connected to a stationary engine on a test-bed (or on a vehicle). There are significant differences between these two scales of tests and modelling techniques often help to bridge that gap. Examples of such studies, where modelling is used, can be found in many papers (e.g. Kandylas et al., 1999; Ahmadinejad et al., 2008).

The need to assign values for the effective diffusivity of reactants in the catalyst layer is generally understood, but there is a lack of data to provide confidence in the method/values used, especially in the area of monolith wall temperatures. Models also vary in complexity, ranging from single channel to multi-channel monolith structures that are modelled as a continuum. Some of the models have many tuneable parameters and the link between cause and effect is not transparent. Many researchers develop their own in-house codes, or sometimes they adapt commercially available CFD codes such as fluent to model the structures. In a number of instances, the work has been supported by transient experimental studies, e.g. Nibbelke (1998) and Harmasen et al. (2001), using small samples of catalyst in bench top studies.

At present, when trials are performed using a Full-scale DOC on a real engine, then the overall performance of the converter is evaluated and an opportunity is often lost to obtain a deeper understanding of the performance of the converter on-line. This arises because, in these tests, there is often a lack of information about temperature profiles along the length of the DOC. Hence, an opportunity is lost to test the validity of mathematical codes which could then be used to help design the DOC for a specific commercial application. For example, when looking at work by Sampara et al. (2008), a description is provided of both small scale experiments (with 19 mm and 38 mm diameter samples) and also of experiments using a Full-scale DOC connected to an engine, and the work is supported by modelling. Although these results are most helpful, the emphasis is placed very much on comparing the conversion across the reactor (model versus experiment), and in comparing the shape and alignment (model and experiment) of the light-off plots. Also, these experiments were done at pseudo-steady state conditions so they cannot test the transient aspects in a simulation code. Likewise, in another example paper (Kim and Kim, 2009) on the modelling of DOCs, comparisons between measured experimental values and modelled data are made by looking at changes in concentration across the DOC as a function of gas inlet temperature. There is some reference to the measurement of substrate temperature but this is not discussed in detail, although an average substrate temperature versus a gas inlet temperature does feature in one of the comparative plots (measured versus modelling).

This lack of data on experimental variations in substrate temperature also applies to developments of catalytic converters for gasoline fuelled vehicles. In a review of the literature in McCullough et al. (2004), the authors conclude that '*the accuracy of the emissions predictions is limited by the surface reaction rate equations*'. This they state: '*is, in part, due to the complexity of the conversion process and the fact that the reaction rates vary with both gas composition and the catalyst formulation*'. They place emphasis on the importance of obtaining rate expressions for new catalyst formulations, which requires extensive experimental testing, and unfortunately the availability of such data is limited. Then, when discussing the importance of validating the computer model used (to simulate or interpret data), they acknowledge that such models can be used to estimate various parameter such as '*internal gas concentrations and substrate temperatures, which are difficult or even impossible to measure using a conventional test apparatus*'.

The challenge of measuring the substrate temperatures along the axis of a Full-scale DOC that is connected to a live engine has already been described in two of our earlier papers on this theme (Kolaczkowski et al., 2012; Ye et al., 2012). So, in this current paper, it is shown how this technique can be used with other on-line measurements to interpret what is happening in a DOC during an NEDC test cycle. This test cycle is