

## Spatial Resolution of Reactant Species Consumption in Diesel Oxidation Catalysts

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### Introduction

There is growing interest in more widespread use of lean-burn engines, such as the diesel engine, in passenger vehicles. Such a switch will lead to decreased fuel consumption and a coincident decrease in CO<sub>2</sub> emissions, both of increasing concern. The remaining issue is NO<sub>x</sub> and particulate emissions. For this reason, there has been substantial research directed at diesel and lean-burn gasoline exhaust emissions control catalysts. A common catalyst in many proposed systems is a diesel oxidation catalyst (DOC). DOCs perform a range of functions in an integrated emission control system, including oxidation of exhaust hydrocarbons and carbon monoxide, and conversion of NO to NO<sub>2</sub>. The latter oxidation reaction is desired for optimum performance in downstream devices; the 'fast' reaction pathway in selective catalytic reduction catalysis requires 1:1 NO:NO<sub>2</sub>, NO<sub>2</sub> is trapped more readily by NO<sub>x</sub> storage/reduction catalysts and soot is burned at a lower temperature with NO<sub>2</sub> as the oxidant relative to O<sub>2</sub>. Furthermore, DOCs can be used to initiate activity in downstream devices by providing heat to the exhaust gases via exothermic reactions with added fuel or fuel-derived hydrocarbons.

While there are DOC global reaction mechanisms available, validation of these mechanisms is traditionally accomplished by simply predicting/matching the measured catalyst out species compositions. A more challenging indicator of the global reaction mechanism validity is the accurate prediction of the species concentration profiles along the catalyst channel. The overall scope of this project is directed at evaluating DOC global mechanisms based on measured species profiles for steady-state reactor experiments. This presentation will show experimentally measured distributions of gas-phase species along the axial direction of the catalyst with focus on characterizing key DOC reactions.

### Materials and Methods

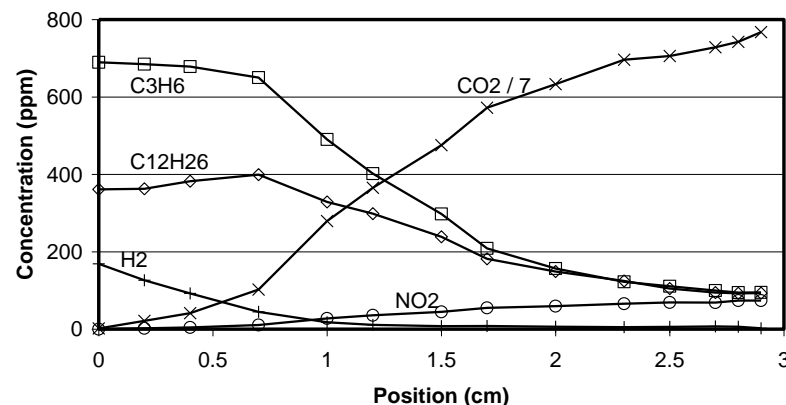
Spatially resolved capillary-inlet mass spectrometry (SpaciMS) [1,2] was used to measure the gas-phase species within a model, monolith-supported Pt-Pd/Al<sub>2</sub>O<sub>3</sub> model diesel oxidation catalyst. Spatially resolved measurements were made using a Hiden mass spectrometer during temperature ramps with the capillary positions fixed, as well as at a constant upstream temperature and the capillary pulled to different positions. Most experiments included a mixture of CO, H<sub>2</sub>, C<sub>3</sub>H<sub>6</sub>, C<sub>12</sub>H<sub>26</sub> and NO, in 5% H<sub>2</sub>O, 10% O<sub>2</sub> and a balance of N<sub>2</sub>. Outlet concentrations were also monitored with a MKS 2030 FTIR.

### Results and Discussion

In the full mixture of reactants described above, the data demonstrate that H<sub>2</sub> and CO are oxidized prior to C<sub>3</sub>H<sub>6</sub> which is oxidized prior to C<sub>12</sub>H<sub>26</sub>. At higher flowrates, back-to-

front oxidation was observed for these species, while at lower flow, H<sub>2</sub> and CO followed the back-to-front ignition pattern, but the C<sub>3</sub>H<sub>6</sub> and C<sub>12</sub>H<sub>26</sub> began oxidizing about mid-way through the sample. There was not consistent, clear evidence depicting a difference in the oxidation of H<sub>2</sub> or CO as a function of temperature. Experiments with fewer reactants demonstrated that NO outlet concentrations were suppressed when C<sub>3</sub>H<sub>6</sub> was added. Adding CO or H<sub>2</sub> to this mixture resulted in little change in the negative effect of C<sub>3</sub>H<sub>6</sub>.

An example of data obtained along the catalyst length at a constant inlet temperature, and after steady-state had been reached, is shown in Figure 1. The H<sub>2</sub> and CO, based on a CO<sub>2</sub> mass balance, are oxidized within 1.5 cm of the inlet, while the hydrocarbons begin oxidizing after 0.75 cm from the inlet and continue to the outlet. Such measurements provide key data for model validation as well as measures to compare as a function of catalyst aging.



**Figure 1.** Species concentration data measured at different positions within the catalyst. Data were obtained at an inlet temperature of 248°C, and inlet concentrations of 160 ppm H<sub>2</sub>, 425 ppm C<sub>12</sub>H<sub>26</sub>, 665 ppm C<sub>3</sub>H<sub>6</sub>, 730 ppm CO, 130 ppm NO 5% H<sub>2</sub>O, 10% O<sub>2</sub> and a balance of N<sub>2</sub>.

### Significance

With increasingly stringent emissions regulations, more robust and efficient exhaust aftertreatment catalysts are being developed. For lean-burn engines, these are just now being introduced, and a common catalyst to these systems is an upstream oxidation catalyst. The results to be presented are being used to build models of these systems, for engine control strategies as well as catalyst development.

### References

1. Choi, J.S., Partridge, W., and Daw, C.S., *Appl. Catal. A: General*, 293, 24 (2005).
2. Choi, J.S., Partridge, W., and Daw, C.S., *Appl. Catal. B: Environmental*, 77, 145 (2007).