Anomalous behavior of NO oxidation over DOCs aged under laboratory and real-world conditions

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Introduction

Diesel oxidation catalysts (DOCs) are used in most diesel exhaust aftertreatment systems to meet exhaust emission regulations. Precious metals platinum (Pt) and palladium (Pd) are the primary active DOC components [1]. The key functions of DOCs are to oxidize: hydrocarbons (HC) and CO emissions from the engine, HC injected to raise the temperature of aftertreatment system, and NO to NO_2 for the oxidation of soot and the enhancement of the SCR reactions [2-4]. The latter function of DOC, oxidation of NO to NO_2, is becoming increasingly critical for the operation of aftertreatment systems. We have observed a complex behavior of this reaction which may show a hysteresis depending on the operation conditions and the history of the catalyst. In this study, we will describe the details of the behavior and the potential factors that lead to such behavior.

Materials and Methods

The experiments were run in a fixed bed continuous-flow reactor with monoliths (0.25 in. x 1 in.) cut from the DOCs of interest. The experiments were conducted with a GHSV of 60,000h^{-1} under ambient atmospheric pressure. The gas composition was 2.5% H_2O, 5% O_2, 5% CO_2, 200 ppm NO balance helium. In this study, catalytic behavior was probed using the oxidation of NO to NO_2. The NO oxidation was quantified using a series of isothermal, steady-state measurements.

Results and Discussion

The activity of the lab-aged catalyst at a given temperature during a ramp up in temperature was different than at that same temperature during a ramp down in temperature (Figure 1(a)). However, DOCs that were exposed to prolonged real-world exhaust conditions did not show measurable differences in oxidation activity as shown in Figure 1(b). Factors responsible for such difference in oxidation activity were explored by perturbing the precious metal oxidation state and supported by various treatments such as reduction and sulfation/desulfation treatments. We identified that the redox state of the precious metal played a crucial role in determining the DOC oxidation activity.

It is postulated that the differences in the initial oxidation state of the PGM and the rate of change in the redox state of the PGM during temperature ramp up and ramp down leads to the observed hysteresis behavior. In this presentation we will show detailed analyses of the factors responsible for such hysteresis behavior. We will also discuss ways of minimizing the behavior and maintaining DOC performance.

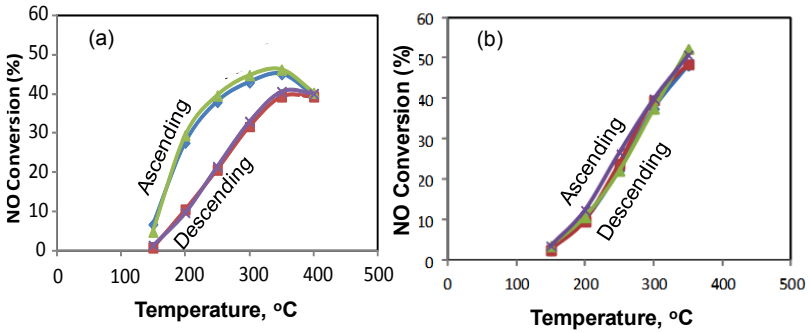

Figure 1: Steady-state NO oxidation performance measured during increasing and decreasing temperature ramps (a) lab aged DOC with no sulfur (b) field-aged DOC with sulfur.

Significance

These findings could pave the way for DOC performance management strategies and DOC catalyst formulation improvements.

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References

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