Catalyzed soot filters for diesel vehicle emission control

Yuejin Li
BASF Corporation, Iselin, New Jersey 08830 USA
yuejin.li@basf.com

Introduction

Worldwide emission regulations have become increasingly stringent for the past decades. For example, for European diesel passenger cars, the emission standards for HC+NO_x and particulate have been decreased by a factor of 6 and 30, respectively. To meet increasingly demanding emission targets, advance catalyst technologies are essential. Catalyzed soot filter (CSF) has been widely used in Europe as an effective device for particulate control for diesel passenger cars since 2005. In addition to its particulate function, CSF has become increasingly important in converting gaseous pollutants in advanced emission control systems. Depending on the catalyst system configuration, CSF can be used to convert additional hydrocarbon and CO after a diesel oxidation catalyst, to oxidize NO to NO_2 for filter regeneration or for maximizing the NO_x conversion of a down-stream SCR catalyst, or to clean up H_2S generated by a lean NO_x trap catalyst. Soot filter is also widely used as a carrier to house urea SCR catalyst to simultaneously control NO_x and particulate emissions.

This presentation focuses on the catalysis side of CSF, specifically on the oxidation of NO to NO_2 over Pt-based CSF and on the use of NO_2 to regenerate soot filter. For filter regeneration, I will focus on the phenomenon of multiple NO to NO_2 turnovers during NO_2+C reaction. For catalytic NO oxidation, I will discuss the effects of Pt/Pd ratio, aging temperature, hydrocarbon species, Pt crystallite size on NO conversion and turnover frequency.

Materials and Methods

Silicon carbide wall-flow filters (34x34x150mm square segments) were used for the experiments. A SiC filter substrate was loaded with catalytic components using a conventional washcoating process. For NO oxidation experiments, a series of Pt/Pd catalysts was used with Pt/Pd weight ratios of 1:0, 10:1, 8:1, 4:1, and 2:1. Pt and Pd were sequentially impregnated on an Al_2O_3-based support by the incipient wetness technique. The total washcoat loading was maintained at 18 g/liter, and the metal loading was between 0.88 to 1.06 g/liter.

NO oxidation over CSF was measured using a lab reactor with a feed consisting of 75 ppm HC (15 ppm C_3H_6, 60 ppm $C_{10}H_{22}$, based on C1), 100 ppm NO, 7% H_2O, 10% O_2, balance N_2 at GHSV=20,000 /h. The reactor temperature was ramped from 100 to 420 ^oC at 15 ^oC/min. The catalysts were tested as fresh and after hydrothermal aging (600 ^oC/1h, 750 ^oC/5h and 850 ^oC/5h). For filter regeneration, a CSF segment was first loaded with 3.5g/l diesel engine soot, and the soot loaded filter was regenerated using a lab reactor at 400 ^oC with a feed consisting of 230 ppm NO, 10% O_2, and 5% H_2O in N_2. CO_2 formation was used to calculate the amount of carbon burned as a function of time. The overall regeneration efficiency was also obtained by weighing the filter before and after the regeneration.

Results and Discussion

Fig.1 shows the regeneration of a soot loaded filter using NO_2 generated on the CSF. At 400 ^oC, the ΔP was first quickly decreased and then followed by a gradual decrease. The cumulative carbon burned, however, increased with time uniformly. CO_2, a product of C+NO_2 reaction, can be used as a measure for NO_2 consumption. By counting the amount of NO_2 consumed (through CO_2 formation) and the amount of NO_2 observed at the CSF outlet, one can conclude that NO has gone through more than one turnover during the regeneration process, and the total NO conversion has well passed its thermodynamic equilibrium (52% at 400 ^oC).

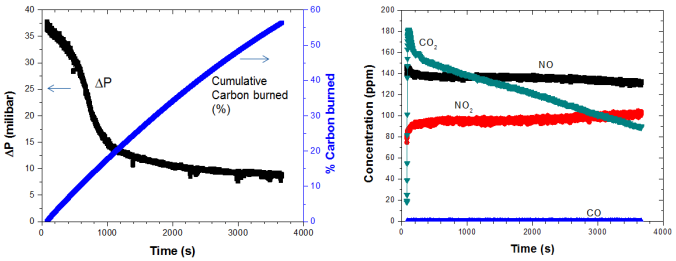

Figure 1 Regeneration of soot loaded filter using NO_2 at 400 ^oC.

Fig.2 shows that CSF treated at 600 ^oC (degreened) has the highest NO conversion, and hydrothermal aging at 750 and 850 C results in increasingly sever deactivation. Interestingly, a fresh catalyst is far less active than a 750 ^oC aged catalyst. Pt/Pd CSF catalysts behave similarly in this regard. However, their relative stabilities against aging are different. Incorporating Pd in Pt decreases the degreened activity significantly and the 750 ^oC aged activity slightly. The activity after 850 ^oC aging, however, is higher on Pt/Pd CSFs. For overall performance, a Pt/Pd CSF with a high Pt content is preferred.

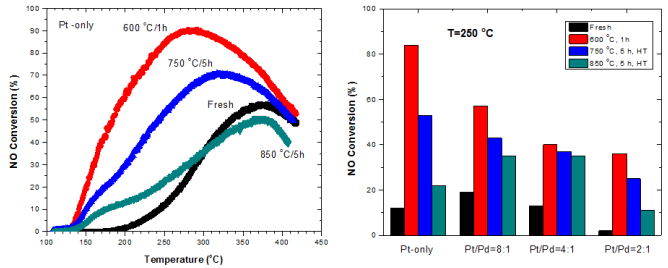

Figure 2 Left: NO conversion on Pt-CSF as a function of aging condition and temperature. Right: NO conversion at 250 ^oC as a function of Pt/Pd ratio and aging condition.

Significance

For a DOC+CSF system, NO oxidation on CSF is important for passive filter regeneration because multiple NO turnovers on CSF make the regeneration more efficient. For a DOC+CSF+SCR system, NO_2 generation on CSF enhances the SCR efficiency, and to achieve stable NO oxidation, a Pt rich Pt/Pd CSF with a right particle size is needed. Increasingly, incorporating catalytic functions into a particulate filter is a cost effective way to meet a number of emission targets simultaneously.