Dual SCR Catalyst System for Lean NOx Reduction

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Introduction

Low-cost lean NOx aftertreatment is one of the main challenges facing high-efficiency gasoline and diesel engines operating with lean exhausts. While there are many candidate technologies, they all offer tradeoffs. We have investigated a multi-catalyst Dual SCR aftertreatment system that is capable of obtaining NOx reduction efficiencies of greater than 90% under lean conditions, without the use of platinum group metals (PGMs) or urea injection into the exhaust. This Dual SCR approach uses a Ag HC-SCR catalyst followed by an NH_3-SCR catalyst. In bench reactor studies from 150°C to 500°C with modest C/N ratios, NOx reacts over the first catalyst to predominantly form nitrogen. In addition, it also forms ammonia in sufficient quantities to react on the second NH_3-SCR catalyst to improve overall performance. The operational window and the formation of NH_3 are improved in the presence of small quantities of hydrogen (0.1-1.0%). The response of the system to other factors such as exhaust oxygen content and space velocity has also been explored. This approach to lean NOx reduction is well matched to low-temperature diesel combustion where HC emissions can be significant and is a potential enabler to satisfy Tier2/Bin5 and Tier2/Bin2 emissions standards.

Materials and Methods

The bench reactor for these experiments has two heated zones, one for vaporization of liquid HC fuel (e.g., iso-octane, dodecane) which is injected into the main feed stream in the second zone holding the catalyst(s) with a temperature ramp between 150°C and 550°C. The HC-SCR catalyst is 2 wt% silver washcoated onto Al_2O_3 in beads or a monolith. The second NH_3-SCR catalyst is a Fe zeolite coated monolith. Standard reactants of our simulated exhaust feed include NO, NO_2, CO, C_3H_8, C_7H_8, C_8H_{18}, $C_{12}H_{26}$, and 0-8% O_2, 4% CO_2, 6% H_2O, with balance N_2. Typically a C/N ratio of 3:1 or 6:1 was used, where the C is from HCs and the N from NOx. Bench analyzers, a GC, and both an electron-excited and a chemical ionization mass spectrometer measured products. Typical space velocities were 27,000 to 50,000 h^{-1}.

Results and Discussion

Catalyst testing for this study was done in three phases. First, the NOx conversion and product distribution of the silver HC-SCR catalyst was measured for a range of lean conditions while varying a HC, e.g. dodecane. A typical NOx conversion for conditions with C/N=3 is shown in the lower curve in Fig. 1. In those studies moderate amounts of ammonia were produced that could react with the remaining NO on a second catalyst. Then the performance of the NH_3-SCR catalyst was measured under conditions like those found exiting the silver catalyst. Finally, both catalysts were measured under the same conditions with a much higher overall conversion observed, as shown in the upper curve in Fig. 1. In this case, the improved performance is the result of the reaction of N-containing species, including ammonia, over the second catalyst. For the same use of HC and H_2 we find the overall NOx conversion for the combined catalyst system is increased over a wide temperature range. Varying oxygen content was not important until it fell below 2%, reducing NOx conversion.

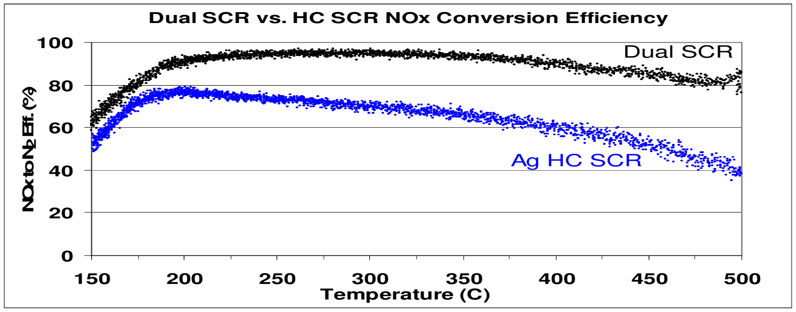

Figure 1. HC-SCR vs. the Dual SCR NOx Reduction Approach. 27000 h^{-1}, C/N=3, 1% H_2, 8% O_2, 4% CO_2, 6% H_2O 140 ppm C_3H_8, 180 ppm NO/NO_2(1:1), 50ppm $C_{12}H_{26}$.

In order to optimize the performance of the second catalyst for NH_3-SCR, a NH_3/NO ratio of 1:1 is preferred under most conditions. In Fig. 2 the impact of increasing HC amount on NOx conversion is shown in a plot vs. C/N ratio at 300°C. As the C/N ratio increases, the amount of NH_3 exiting the Ag catalyst increases and the NOx amount decreases. Most of the unreacted NOx appears as NO. The NH_3 and NO amounts are equal for C/N=3, near the same C/N ratio as the peak NOx conversion to N_2. This is a lower C/N ratio than found for many catalysts that often use C/N=6 or higher. The Dual SCR system uses NH_3 to further improve NOx conversion and keep the fuel economy penalty to a minimum for NOx control on a vehicle.

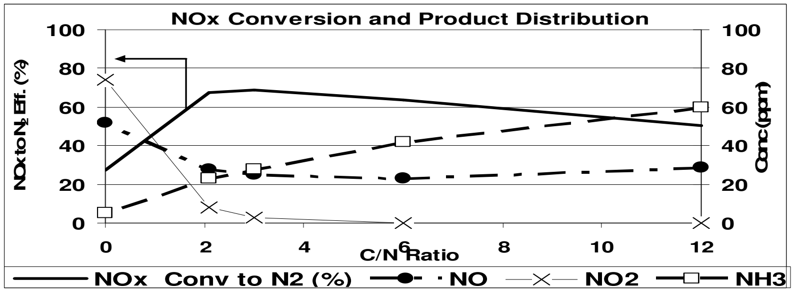

Figure 2. Ag HC-SCR NOx Product Dependence on C/N. 300°C, 27000 h^{-1}, 1% H_2, 8% O_2, 4% CO_2, 6% H_2O, 140 ppm C_3H_8, 180 ppm NO/NO_2 (1:1), with variable HC, $C_{12}H_{26}$.

Significance

In lean exhaust conditions the overall NOx conversion of a silver HC-SCR catalyst is enhanced when followed by an NH_3-SCR catalyst. This is accomplished by the appropriate control of hydrocarbons and hydrogen in the feed to provide sufficient amounts of ammonia such that the second catalyst further raises the overall NOx conversion to N_2.