The Effect of Oxygen Concentration in the Regeneration Phase on the Performance of a NOx Storage/Reduction Catalyst

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Introduction

NO_X storage/reduction (NSR) catalytic systems are being used and developed for lean-burn engine exhaust-gas NO_X emissions reduction to meet existing and upcoming regulations in Europe and North America. Unlike gasoline-exhaust catalytic converters, which operate consistently under nominally stoichiometric conditions, NSR catalyst systems operate cyclically between two distinct phases. During the lean phase (normal diesel conditions), NO is oxidized and stored on the surface of the catalyst as nitrites and nitrates. The second phase is the rich phase where reductant is introduced and the trapped NO_X is released and reduced on the catalyst [1].

Materials and Methods

The NSR catalyst used in this study is composed of $Pt/Ba/Al_2O_3$ supported on a monolith. A standard tube-reactor was used to examine the effects of including O_2 in the rich phase of NSR catalyst regeneration. Experiments were directed towards a) evaluating the effects of residual O_2 in the exhaust during the regeneration phase and b) NO_X reduction optimization as a function of O_2 rich-phase concentration. The temperatures studied range from 200 to 550°C. A standard 60-second, 300 ppm NO lean phase was used for each test in this study. At each temperature, the amount of O_2 or reductant was varied in a 5-second rich-phase stream, until steady-state cycle-to-cycle conversions were observed. The experimental concentrations of reductant and O_2 used in the rich phase for one example set of tests are shown in Table 1. Note the net amount of reductant, assuming complete reaction of O_2, is identical for Rich 1 and 3, and 2 and 4.

Table 1. Rich-Phase Reductant and Oxygen Concentrations

Reductant	Rich 1	Rich 2	Rich 3	Rich 4
H_2	0.675%	1.050%	0.375%	0.450%
CO	1.125%	1.750%	0.625%	0.750%
O_2	0.30%	0.90%	0.00%	0.00%

Results and Discussion

Results obtained at 375°C are shown in Figure 1. The plot shows the outlet reactor NO_X concentration over one 65 second cycle. It was assumed that addition of O_2 would result in some reductant being consumed via oxidation, and therefore less would be available for nitrate reduction and catalyst regeneration. A coincident effect would be a temperature rise on the sample from the exothermic oxidation reaction. The experiments were targeted to answer how this temperature rise might affect performance.

To investigate this O_2 effect, 0.3% O_2 was added to 1.8% reductant to see how a small increase in rich-phase O_2 would affect catalyst performance (Rich 1) in comparison to 1.2% reductant (Rich 3). Note, these two mixtures contain the same amount of reductant if all of the O_2 is consumed by reductant. As shown, a slight increase in trapping performance was observed in comparison with the O_2-containing stream. Similarly, with an increased reductant concentration of 2.8% and an O_2 concentration of 0.9%, (Rich 2), better performance was obtained in comparison to 1.0% reductant (Rich 4). The data for Rich 1 and Rich 2 showed similar NO_X outlet concentrations throughout the cycle. A maximum in improvement was observed as a function of O_2 added. The inclusion of rich phase O_2, to a specific degree, suggests an increase in catalyst performance. These data were reproducible and experiments conducted at lower temperatures followed the same data trend. However, at higher experiment-starting temperatures, the addition of even the small amounts of O_2 resulted in decreased performance. There were also observed differences in NH_3 and N_2O byproduct generation during the regeneration phase with O_2 addition. These results will be related to the temperature changes associated with exothermic oxidation as well as the effect of O_2 on NO_X release during the rich phase.

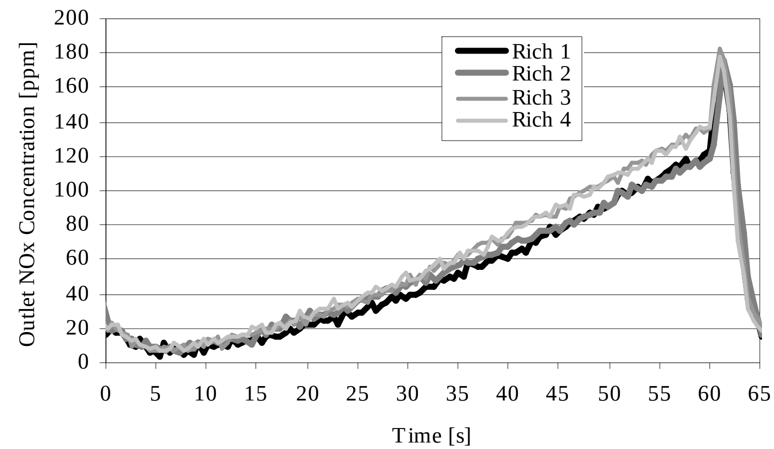

Figure 1. Outlet NOx concentration during a single cycle after steady cycle-to-cycle performance was reached, 375°C.

Significance

The data presented above supports the idea that the addition of small amounts of O_2 in the regeneration phase results in an increase in the NOx storage ability of the catalyst. These results can be used to optimize NSR catalyst performance.

References

1. W.S. Epling, L.E. Campbell, A. Yezerets, N.W. Currier, and J.E. Parks, *Catal. Rev.* 46, 163(2004).