

Assessing a Commercial Lean NO_x Trap Performance via Spatiotemporal Species Profile Measurements

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Introduction

The Lean NO_x Traps (LNTs), also known as NO_x Storage-Reduction (NSR) catalysts, are a promising NO_x abatement technology to meet ever more stringent future nitrogen oxides emission standards for diesel vehicles [1]. To circumvent the problem of inefficient catalytic NO_x reduction under the net-oxidizing (lean) environment of diesel exhaust, LNTs combine a NO_x storage component (alkali or alkaline-earth metal) to the conventional three-way catalyst [2]. The LNT stores NO_x during normal lean diesel operation (storage step) and releases and reduces the stored NO_x during intermittent short rich excursions (regeneration step) [2]. The LNT operation is thus both cyclic and integral in nature, which makes its fundamental understanding using the traditional reactor inlet/outlet measurement approach difficult [2]. In this respect, the ability to measure local species profiles inside a working catalyst with high temporal resolution is highly desirable.

In this paper, we report LNT research employing a new experimental approach, i.e., measurement of spatiotemporal species profiles inside a working catalyst [3]. One novelty of this work is to have addressed a commercial LNT sample whose composition is much more complex than that of the model catalysts extensively reported in the literature. We have evaluated the reaction characteristics of the selected commercial LNT on a bench flow reactor by monitoring axial species distributions with Spatially-resolved capillary-inlet Mass Spectrometry (SpaciMS). Special attention was given to NO_x storage distribution, oxygen storage, reductant evolution, and the impact of sulfation and desulfation on different LNT reactions.

Materials and Methods

A commercial LNT (97 cells per cm²) has been provided by Umicore through U.S. DOE CLEERS (Crosscut Lean Exhaust Emissions Reduction Simulations: <http://www.cleers.org>) LNT Focus Group [4]. An elemental analysis showed Al, Ba, Ce, Zr, Pt, Pd and Rh to be the major components. We evaluated a 2.1-cm diameter and 7.4-cm long core sample in a horizontal quartz reactor heated by an electric furnace. The experiments were performed in a cyclic mode (60-s lean/5-s rich) at 230, 325 and 500 °C with a space velocity of 30000 h⁻¹. The lean phase gas mixture contained 300 ppm NO, 10% O₂, 5% H₂O, 5% CO₂ and N₂ balance, while the rich phase gas mixture contained 1.4 or 3.4% reductant (H₂, CO or H₂/CO), 5% H₂O, 5% CO₂ and N₂ balance. After a stationary cyclic operation was established, the gas composition was analyzed by SpaciMS at different catalyst locations.

Results and Discussion

The obtained species profiles clearly show where NO_x is stored at different temperatures and permit meaningful performance comparisons. For example, even though NO_x

storage capacities are quite equivalent at 325 and 500 °C based on the outlet lean-phase breakthroughs, a much longer portion of the LNT was necessary at 500 °C (see Fig. 1). The data also indicate the important role of the thermal desorption at high temperatures resulting in increasing and broadening NO_x spike along the catalyst during the rich phase.

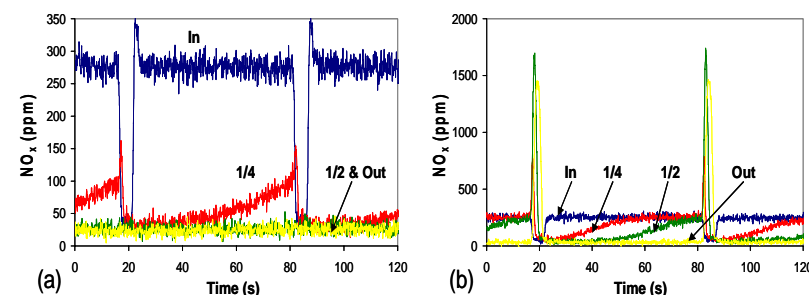


Figure 1. Spatiotemporal NO_x profiles at (a) 325 °C and (b) 500 °C during 60'' lean/5'' rich operation of a commercial LNT (3.4% H₂ as reductant).

To quantify the oxygen storage capacity (OSC) of the LNT, we performed the same cycling experiments but without NO during the lean phase. Consistent with the elemental analysis indicating significant Ce and Zr presence, the studied sample showed large dynamic OSCs. As a result, significantly higher reductant than that necessary to remove only the stored NO_x, was necessary. However, as NO_x storage is more localized at the LNT front whereas oxygen storage is more evenly distributed, it would be possible to optimize the quantity and duration of reductant in order to maximize the NO_x removal performance and minimize the reductant consumption. Another notable reductant chemistry involved in LNT operation in the presence of CO is the water-gas shift (WGS) reaction. The studied LNT showed significant WGS activity.

After describing in detail intra-catalyst NO_x and reductant chemistry, we will discuss the effect of sulfation and desulfation on the different reactions in this presentation.

Significance

The spatiotemporal species profiles obtained in this work provide localized chemistry information for a commercial LNT under realistic conditions. This localized information will be useful in LNT model development, catalyst design and operation.

References

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