

## NO<sub>x</sub> removal over a double bed NSR-SCR reactor configuration

L. Lietti, P. Forzatti \*

Dipartimento di Energia, Centro NEMAS—Nano Engineered Materials and Surfaces,  
Politecnico di Milano - p.za Leonardo da Vinci, 32 - 20133 Milano

\*pio.forzatti@polimi.it

### Introduction

NO<sub>x</sub> Storage-Reduction (NSR) catalytic systems and urea-SCR are both considered viable solutions for the NO<sub>x</sub> removal under lean conditions<sup>[1]</sup>. In NSR systems (e.g. Pt-Ba/Al<sub>2</sub>O<sub>3</sub>) the removal of NO<sub>x</sub> is realized under cyclic conditions, by alternating long lean periods during which NO<sub>x</sub> are adsorbed on the catalyst with short excursions under rich conditions during which the stored NO<sub>x</sub> are reduced to N<sub>2</sub>. On the other hand in the urea-SCR technique the NO<sub>x</sub> reduction is accomplished by injecting urea (a precursor of NH<sub>3</sub>) in the exhaust gases. Although urea-SCR is preferred for heavy trucks and mini-van, NSR catalysts are cheaper for small engines<sup>[1]</sup>. A drawback related to NSR systems is the emission of undesired NH<sub>3</sub> during the rich phase, especially at low temperatures<sup>[2]</sup>. A possible solution is to place a SCR catalyst downstream the NSR system, so that NH<sub>3</sub> formed over NSR during the rich phase can be stored on the SCR catalyst, and consumed in the subsequent lean phase<sup>[3]</sup>. This might lead in principle to both a decrease of the NH<sub>3</sub> slip, and a simultaneous increase of NO<sub>x</sub> removal. The potentiality of combined NSR-SCR systems, in which NH<sub>3</sub> formation over NSR is exploited on a SCR catalyst, is considered in this work by using H<sub>2</sub> as a reductant.

### Materials and Methods

A homemade Pt-Ba/Al<sub>2</sub>O<sub>3</sub> (1/16/100 w/w) model catalyst and a Fe-ZSM5 SCR catalyst provided by Zeolyst International (both in the form of powder) have been used in this study. Lean-rich cycles with either a single NSR or SCR catalyst bed or with a double bed arrangement (with the Fe-ZSM5 SCR bed placed downstream the NSR sample) have been carried out at different temperatures. The lean phase consist of a pulse of NO (1000 ppm) in He + 3 % v/v O<sub>2</sub> until catalyst saturation. Then after a He purge the rich phase has been carried out with H<sub>2</sub> (2000 ppm in He). The tests have been performed in a quartz tubular fixed-bed microreactor connected to a mass spectrometer for analysis of reactants and products<sup>[2]</sup>.

### Results and Discussion

Figure 1 compares the behavior of the NSR catalyst upon a lean-rich cycle at 200 °C (Figure 1A and 1B) with that of the NSR+SCR double bed configuration (Figure 1C and 1D). Upon the lean phase the NO<sub>x</sub> breakthrough is seen near 200 s for the NSR catalyst (Figure 1A); then the NO<sub>x</sub> concentration (mostly NO) increases up to the inlet NO concentration. During the subsequent rich phase (Figure 1B) N<sub>2</sub> and NH<sub>3</sub> are detected at the reactor outlet, with NH<sub>3</sub> following N<sub>2</sub> evolution.

In the case of the double bed configuration (Figure 1C and 1D) N<sub>2</sub> is detected also during the lean phase (Figure 1C) and the amounts of NO<sub>x</sub> removed from the gas phase during this phase are higher than those removed in the case of the single NSR catalyst bed. Besides, during the rich operation no ammonia is observed at the reactor outlet (Figure 1D). Hence the results clearly indicates that ammonia released from the NSR catalyst bed during rich operation is stored on the SCR catalyst bed placed downstream: the stored ammonia reacts with NO<sub>x</sub>

exiting the NSR bed during the subsequent lean operation, as pointed out by N<sub>2</sub> formation during NO<sub>x</sub> adsorption (see figure 1C). Accordingly this catalyst bed configuration increases the system efficiency by improving both the selectivity during the rich operation (the SCR catalyst bed located downstream prevents the NH<sub>3</sub> slip) and the NO<sub>x</sub> removal efficiency during the lean operation due to the reaction of NO<sub>x</sub> with NH<sub>3</sub> stored on the SCR catalyst. The benefits of this arrangement is particularly exploited at low temperatures where a significant NH<sub>3</sub> slip is observed from the NSR catalyst and accordingly relevant amounts of NH<sub>3</sub> are stored on the SCR catalyst bed located downstream the NSR sample.

The reactivity of NSR-SCR combined systems in which the NSR catalyst is mixed up with the SCR catalyst in one bed only is presently under investigation. However preliminary results indicate that the benefits of the NSR-SCR arrangement can only be obtained with a two-bed configuration, and results will be presented in forthcoming studies.

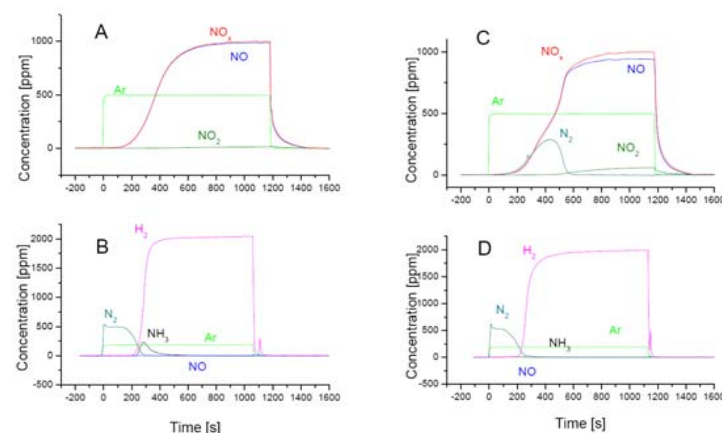


Figure 1- Lean-rich cycles performed at 200°C over the NSR sample (catalyst load: 60 mg, flow rate: 100 cm<sup>3</sup>/min. A: lean operation, B: rich operation) and over the NSR+SCR double bed arrangement (catalyst load: NSR 60 mg, SCR 60 mg, overall low rate: 100 cm<sup>3</sup>/min. C: lean operation, D: rich operation).

### Significance

The rationalization of the behavior of NSR-SCR combined catalyst beds has been attained, which is helpful in the understanding the catalytic behavior of NSR catalysts and which is relevant for the development of improved NO<sub>x</sub> abatement systems.

### References

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