

## Dynamic Behavior of Fe-Zeolite Urea-SCR Catalysts

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### Introduction

Selective Catalytic Reduction (SCR) of NO<sub>x</sub> with urea is one of the primary candidate technologies for meeting the increasingly stringent diesel exhaust emission regulations worldwide. Several types of metal-exchanged zeolite catalysts have been shown to have excellent hydrothermal stability and steady-state NO<sub>x</sub> conversion over a broad range of diesel exhaust conditions, with Fe- and Cu-zeolites being the two leading candidate groups [1,2]. However, realizing these high conversion efficiencies in the practical applications requires detailed understanding of the key individual functions contributing to the overall NO<sub>x</sub> reduction performance of these urea-SCR catalysts, including their capability to catalyze in-situ oxidation of NO and NH<sub>3</sub>, as well as their NH<sub>3</sub> storage and release characteristics [2]. The latter factor is often responsible for large differences between the commonly reported steady-state conversion values, and real conversion efficiencies observed in practice, due to rapidly varying temperature, gas flow rate, and exhaust gas composition. In this study, dynamic responses of several key functions of a commercial Fe-zeolite SCR catalyst were individually characterized over a broad range of experimental conditions relevant to automotive applications (temperatures, flow rate, NO<sub>2</sub>/NO<sub>x</sub> and NH<sub>3</sub>/NO<sub>x</sub> ratio) and related to the NO<sub>x</sub> conversion efficiency.

### Materials and Methods

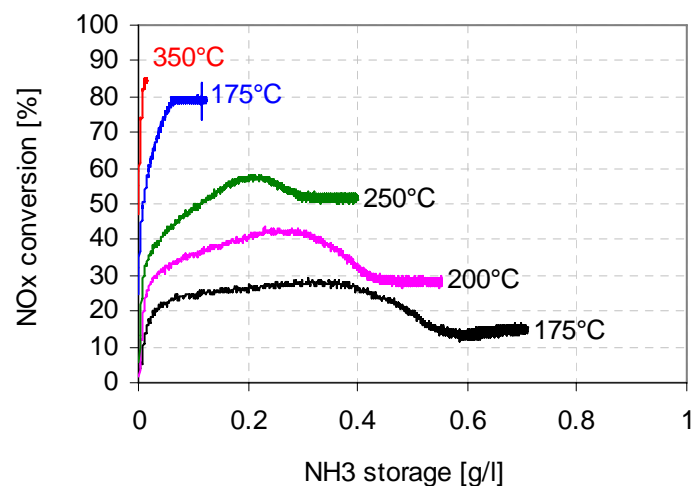
A 4-step test experimental protocol, developed at Cummins and used in this study, provides independent measurements of a variety of SCR catalyst functions. These include dependence of the instantaneous NO<sub>x</sub> conversion on NH<sub>3</sub> coverage, as well as a number of steady-state characteristics, for example NO and NH<sub>3</sub> oxidation. A commercial Fe-zeolite catalyst coated on cordierite honeycomb monolith, with cell density of 300 cells/in<sup>2</sup>, supplied by Johnson Matthey, was cut in to 2.5 cm diameter and 7.6 cm long cylindrical samples and is used in this experimental work. Its performance was investigated in the temperature range of 150-600°C at a fixed space velocity (GHSV) of 40,000 h<sup>-1</sup>. A typical reaction mixture contained 10 vol% O<sub>2</sub>, 8 vol% CO<sub>2</sub>, 7 vol% H<sub>2</sub>O, 200 ppm NO<sub>x</sub> (NO<sub>2</sub>/NO<sub>x</sub>=0 to 1, when used), 200ppm NH<sub>3</sub> and balance N<sub>2</sub>. The concentrations of various exhaust species were measured by MKS FT-IR 2030 MultiGas Analyzer.

### Results and Discussion

The studied catalyst was found to have a large NH<sub>3</sub> storage capacity, such that it took tens of minutes from the beginning of NH<sub>3</sub> dosing to reach steady-state conditions at lower temperatures. During that period, instantaneous NO<sub>x</sub> conversion efficiency was changing substantially as a function of ammonia coverage, as shown in Figure 1. At relatively low coverage levels, NO<sub>x</sub> conversion increased with the coverage, as expected. However, at higher coverage levels, realized at low temperatures, the conversion declined when approaching dynamic saturation. Independent measurements of NO oxidation showed that the

studied catalyst has appreciable ability to generate NO<sub>2</sub> in-situ under the reaction conditions, which is beneficial when the NO<sub>2</sub>/NO<sub>x</sub> ratio in the inlet gas composition is below the optimal value of 0.5. The findings in this work suggest that the in-situ NO oxidation function is inhibited at higher levels of NH<sub>3</sub> coverage, resulting in the decline of SCR efficiency at higher NH<sub>3</sub> coverage levels, as illustrated by Figure 1.

The employed experimental protocol also allowed us to distinguish between the “dynamic” NH<sub>3</sub> storage capacity reached at steady-state SCR conversion conditions, and “total” NH<sub>3</sub> storage capacity, achievable at the same conditions in the absence of the SCR reaction. The difference between the dynamic and total NH<sub>3</sub> capacity can be very substantial, depending on the rate of the SCR and NH<sub>3</sub> oxidation reactions. In this work we have also shown that pre-saturating the catalyst to the optimal level of NH<sub>3</sub> storage at a given set of conditions allows us to reach the best NO<sub>x</sub> reduction efficiency more rapidly.



**Figure 1.** NO<sub>x</sub> conversion dependency on NH<sub>3</sub> surface coverage under “standard” SCR conditions (NO<sub>2</sub>/NO<sub>x</sub>=0) at different temperatures.

### Significance

Function-specific understanding the details of SCR catalyst operation is critical to achieving optimal performance in the practical, transient applications.

### References

1. Grossale, A., Nova, I., Tronconi, E., Chatterjee, D., Weibel, M., *J. Catal.*, 256, 312 (2008)
2. Olsson, L., Sjövall, H., Blint, R. J., *Appl. Catal. B* 81, 203 (2008)