

## Simultaneous soot and NO<sub>x</sub> removal: Experimental investigation over a Cu-zeolite SCR catalyst

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### Introduction

The emerging SDPF technology is becoming more and more attractive, as the tightening emission regulations drive the development of new strategies for the after-treatment of lean exhaust gas (e.g. Diesel exhausts) from mobile sources (cars, trucks, trains and ferries). Within the field of the simultaneous NO<sub>x</sub> and PM abatement, the SDPF, consisting of a NH<sub>3</sub> Selective Catalytic Reduction (SCR) catalyst coated onto a Diesel particulate filter (DPF), is one of most promising technologies [1]. The SDPF affords the reduction of aftertreatment system volume by integrating two unit operations in the same device. It can also alleviate the need of DPF regeneration, since the presence of NO<sub>2</sub>, a better oxidizing agent than O<sub>2</sub>, assists the soot combustion at low temperature. However, it is well known in the literature that NO<sub>2</sub> plays an important role also in determining the deNO<sub>x</sub> activity at low temperatures (below 250°C), due to the so called Fast-SCR reaction [2]: so a competition for NO<sub>2</sub> between soot combustion and NH<sub>3</sub>-SCR chemistry is expected.

This work is particularly focused on three different fundamental aspects: i) understanding the effect of the nature and composition of the oxidizing mixture on the soot combustion, ii) clarifying how the simultaneous presence of NO<sub>x</sub> and NH<sub>3</sub> affect the PM combustion chemistry, and iii) studying the impact of soot on the NH<sub>3</sub>-SCR chemistry, with focus on the key role played by NO<sub>2</sub>.

### Materials and Methods

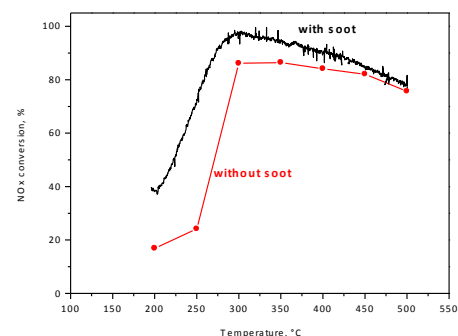
The original commercial Cu-zeolite coated on a ceramic SiC DPF brick was crushed and sieved to a particle size within the 95-106 µm range. The so obtained catalyst powder was then mixed with 10% w/w real soot collected from a Diesel engine run at 3500 rpm and equipped with a DOC. A new reactor was prepared for each run involving soot to guarantee the same load of particulate. NH<sub>3</sub>/NO-NO<sub>2</sub>/O<sub>2</sub> steady-state and transient kinetic runs were performed in a wide and representative temperature range (150 – 500 °C) in order to obtain a NH<sub>3</sub>-SCR reference baseline. PM/O<sub>2</sub>/NO-NO<sub>2</sub> transient runs were also performed over the same catalyst, while PM combustion curves were collected in the 150-700°C temperature window. Finally the PM/NH<sub>3</sub>/O<sub>2</sub>/NO-NO<sub>2</sub> reactive system was investigated under transient conditions in order to study the real scenario. NH<sub>3</sub> adsorption/desorption, NH<sub>3</sub> oxidation and NH<sub>3</sub>-NO-NO<sub>2</sub> tests were performed feeding NH<sub>3</sub>, NO, NO<sub>2</sub>, O<sub>2</sub> with typical concentrations of NO<sub>x</sub> and NH<sub>3</sub> of 500 ppm in N<sub>2</sub> + O<sub>2</sub> (8% v/v) and water (5% v/v). NO/NO<sub>2</sub> and NO<sub>x</sub>/NH<sub>3</sub> ratios varied depending on the type of run. The outlet gas mixture was analyzed by means of a mass spec, a UV analyzer and a ND-IR analyzer devoted to CO<sub>x</sub> detection [2].

### Results and Discussion

The Cu-zeolite based catalyst was studied under both NH<sub>3</sub>-SCR and NO<sub>x</sub> soot combustion conditions. In the soot combustion runs the light-off temperature dropped from

400°C to 250°C upon addition of NO<sub>2</sub> to the feed mixture. The presence of only NO and O<sub>2</sub> in the feed mixture did not affect the light-off temperature and the curve shape during the soot combustion. Accordingly, NO does not interact with the soot even in the presence of O<sub>2</sub>, while on the contrary, NO<sub>2</sub> strongly interacts with soot, promoting a low-temperature soot combustion pathway. NO<sub>2</sub> was also strongly adsorbed on the soot: the related surface compounds reacted with the carbon during the TDP releasing CO<sub>x</sub> as products.

Concerning the NH<sub>3</sub>-SCR activity, our data indicate that, under Standard-SCR conditions, PM did not affect the high activity of the Cu-zeolite in the middle-low T range; only above 400°C the NO slip was enhanced. Also for Fast-SCR conditions (NO<sub>2</sub>/NO<sub>x</sub>=1/2) a limited PM effect was observed, with a small increase of NO above 400°C. On the contrary, the effect of soot on the SCR activity was quite significant in the case of the NO<sub>2</sub>-SCR reacting system. Figure 1 shows for example a strong enhancement of the NO<sub>x</sub> conversion up to 350°C under NO<sub>2</sub>-SCR conditions (NO<sub>2</sub>/NO<sub>x</sub>=1) and in the presence of PM.



**Figure 1.** Comparison between runs w PM and w/o PM, NO<sub>2</sub>-SCR, Q = 172 Ncm<sup>3</sup>/min, O<sub>2</sub> = 8%, H<sub>2</sub>O = 5%, NO<sub>2</sub>=NH<sub>3</sub> = 500ppm, soot load 10% w/w<sub>cat</sub>.

The enhanced deNO<sub>x</sub> efficiency is explained by the partial conversion of the excess NO<sub>2</sub> to NO due to its reaction with soot, so that the optimal NO/ NO<sub>2</sub> = 1:1 ratio of Fast SCR is approached. Furthermore, the reduction of the NO<sub>2</sub> excess also lowered the undesired N<sub>2</sub>O production in the mid-T range (250-400°C)

### Significance

A better fundamental understanding of the mutual interactions between NH<sub>3</sub>-SCR chemistry and PM filtration/combustion can lead to an improved design of SDPFs, thus enabling to reduce volumes and costs of vehicle exhausts aftertreatment systems.

### References

- [1] T.C. Watling, M.R. Ravenscroft, G. Avery, Catalysis Today 188 (2012) 32-41.
- [2] M. Colombo, I. Nova, E. Tronconi, Catalysis Today 151 (2010) 223-230.