

On-Engine Investigation of SCR on Filters (SCRoF) for HDD Passive Applications

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ABSTRACT

A 4-way Heavy Duty Diesel (HDD) emissions control aftertreatment system typically consists of diesel oxidation catalyst (DOC), catalyzed soot filter (CSF), urea based selective catalytic NO_x reduction (SCR) and NH₃ slip control catalyst (AMOX). Incorporating the SCR functionality into the soot filter (SCRoF) has great potential to reduce system costs and package volume/weight.

In this paper, we discuss some of the recent Cu-Zeolite based SCR on filter (SCRoF) developments targeting *Passive* filter regeneration applications. The on-engine investigation of complete DOC+SCRoF+AMOX system focused on three major areas: 1) SCR performance of NO_x conversion efficiency and NH₃ slip under both steady state and transient testing conditions; 2) SCRoF system response to sulfur exposure and subsequent sulfur removal for activity recovery; and 3) Characteristics of filter soot load, pressure drop, and passive soot oxidation in SCRoF.

Overall, the SCRoF system showed comparable NO_x conversion efficiency and sulfur response as the conventional DOC+CSF+SCR+AMOX system. A 92+% cycle cumulative NO_x conversion efficiency was demonstrated in a transient emission testing cycle while the peak tailpipe NH₃ slip was well controlled below 5 ppm. With the tested system, soot load does not adversely affect NO_x reduction. After sulfur exposure, system performance was fully recovered by desulfation at 500°C - often the threshold temperature for passive systems. It is also important to point out that passive soot oxidation (*i.e.* C+NO₂) in an SCRoF will be considerably lower than in a catalyzed soot filter (CSF). Passive regeneration capability is inhibited due to SCR reactions competing for NO₂. In this work, intrinsic mechanisms and possible approaches to further enhance passive soot oxidation are reviewed and discussed.

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INTRODUCTION

In a conventional HDD four way emissions control system, the diesel oxidation catalyst (DOC) converts HC and CO, the catalyzed soot filter (CSF) traps and passively regenerates soot, the selective catalytic reduction (SCR) catalyst removes NO_x, and the ammonia oxidation catalyst (AMOX) converts any NH₃ slip that might come from the SCR catalyst to avoid secondary emissions. By combining the soot filtration function and the SCR NO_x reduction function into one single unit (SCRoF) [1], schematically illustrated in Figure 1, the flow-through SCR substrate(s) can be eliminated and the filter is no longer loaded with precious metals. Total system cost is therefore reduced, as is the package volume and weight. Another key benefit of the SCRoF system is that it has less total thermal mass and can be heated up much faster than the conventional system. This

is of great importance to achieve good NO_x conversion especially in cold start cycles.

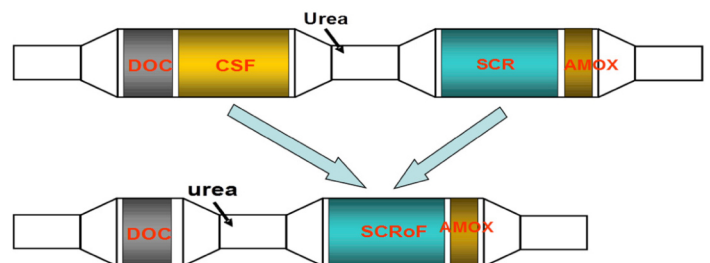


Figure 1. Schematics of the concept of incorporating SCR NO_x reduction function into PM filters.

A number of studies have been conducted at several institutions to look into SCRof as a potential system scenario for HDD application [2, 3, 4, 5, 6]. Recently, researchers at Ford [2] carried out both laboratory and engine-dynamometer tests studying two different copper zeolite SCR formulations coated on several different types of high porosity filter substrates (cordierite and silicon carbide). They concluded that, with the same type of active materials, SCRof was overall able to deliver similar performances as the traditional flow-through SCR system.

Johnson Matthey described in another SAE technical paper [3] the engine evaluation of SCR coated filters. It showed that good NOx conversion efficiency was achievable in the 205-450°C temperature window. After pre-saturation with NH₃, the three tested DOC-SCR-DPF systems converted 82-91% NOx in a hot LA-4 testing cycle. The paper also showed that, compared to a conventional DOC-CSF system, the SCR-DPF system has reduced CO oxidation during regeneration. However the Cu-zeolite SCR catalyst delivered 31-34% NOx conversion across the filter under ANR=1 during filter regeneration. In a later paper [4], the same company studied DOC + SCR-DPF + SCR on a high NOx engine (4.5 g/hp-hr BSNOx, EGR off) and compared the results obtained with a conventional DOC+CSF+SCR system on a low NOx engine (<1.0 g/hp-hr BSNOx, EGR on) 2007MY HDD engine. The SCR-DPF system showed strong evidence of passive soot oxidation at 400°C, however the passive soot oxidation capability was significantly inhibited with urea injection, especially at 300°C.

GM [5] evaluated Cu-zeolite SCR catalyst coated on wall-flow filters on a 6.6L heavy-duty Duramax diesel engine in a test cell. Additionally, a laboratory fixed-bed flow reactor system was used to determine the NH₃ storage characteristics and the SCR performance as a function of soot loading and catalyst aging. It found some reduction of NH₃ storage after soot load in the degreened stage however the impact was minimal after aging.

In addition to the aforementioned experimental work, there also have been quite some efforts to simulate the complex SCRof process, where mathematical governing equations were constructed to couple both reaction and diffusion phenomena [7, 8, 9]. Besides typical SCR reactions, NH₃ adsorption/desorption characteristics and soot load or consumption by NO₂ should be taken into account as well.

An interesting note from the SCRof literature review was that so far all development work documented in open resources focused mostly on active system applications where soot collected in the filter is oxidized at elevated temperatures. The exothermal needed to initiate soot combustion is often realized through in-exhaust fuel injection into a fuel burning DOC. However recent advancement in copper zeolite SCR technology [10], with significantly improved low temperature activity, has permitted OEMs to explore passive system solutions. In such a system, soot regeneration in filters is realized through reaction with NO₂

and active soot combustion is not required. This will lower hardware cost (no fuel injector in exhaust system) and operational cost (less fuel penalty). In this work, the Cu-zeolite SCR catalysts was coated onto a high porosity filter and tested on an engine for HDD passive filter regeneration system applications.

The paper is organized as follows: The Experimental section describes test setup, sampling and measurement techniques, testing protocol, and full size samples used in the study. The Results and Discussion section presents on-engine evaluation results for three major aspects of SCRof for passive HDD applications: SCR NOx conversion performance, sulfur effect that is related to system durability, and passive soot regeneration performance, including filter ΔP and soot oxidation by NO₂. Finally, key findings and a few important remarks are highlighted in the Conclusion section.

EXPERIMENTAL

Engine and Test-Bench

In the experimental work, a prototype 9.3L 2011MY non-road engine together with a motoring electric dynamometer was used. This fully functional heavy-duty test facility is capable of running both steady-state and transient test cycles. It is equipped with constant volume sampling (CVS) and particulate sampling capability, although they were not utilized for this work. In order to simulate beyond 2010MY type engine-out NOx emissions for both on-road and off-road vehicles, an experimental calibration file was used over the course of this study that gave higher BSNOx at 3.2 g/kW-h.

Unless specifically mentioned, all evaluation tests in this work were run with ULSD fuel (nominal 15 ppm or lower wt % sulfur content).

Sample Matrix

Table 1 defines the tested full size parts' dimensions, geometries (CPSI / wall thickness), and respective precious metal loadings. The same DOC+SCRof+AMOX system was used throughout the study, during which the SCRof part had been aged at 650°C for 100h on engine prior to various evaluations. This level of thermal exposure is believed to be well beyond the typical EOL (End of Life) thermal aging requirement for most passive systems. Cu-zeolite SCR catalyst was coated on the high porosity (59%) 300/12 wall flow type of cordierite filters. The actual washcoat loading and sample preparation process was the result of the simultaneous laboratory optimization of multiple parameters: SCR activity, filtration efficiency and backpressure. The SCRof was 12"×12" so the volume ratio of SCR catalyst to engine displacement is 2.4.

As mentioned, this system was designed for passive soot oxidation applications. Therefore sufficient NO₂ concentration is a key factor in order to have good passive soot oxidation. For this purpose, a 50g/ft³ Pt-only 10.5"×6"

DOC was used for NO oxidation. To complete the system, a 12"×2.5" ammonia oxidation (AMOX) catalyst was included downstream the SCRoF to control NH₃ slip.

To compare passive soot oxidation capability with the SCRoF system, a conventional DOC (10.5"×6") + CSF (12"×13.5") layout was also run. As shown in Table 1, the DOC and CSF were loaded with 30 and 5 g/ft³ Pt, respectively. The combined Pt loading of the DOC+CSF system was ~10% less than the DOC+SCRoF system.

Table 1. Full size parts tested in this work.

System	Sample	Size (inch)	Geo-metry	PGM (g/ft ³)
DOC+ SCRoF+ AMOX	DOC	10.5x6	400/6	50
	Cu-SCR on HP filter	12x12	300/12	N/A
	AMOX	12x2.5	400/4	1
DOC+ CSF	DOC	10.5x6	400/6	30
	CSF	12x13.5	200/12	5

System Layout and Sampling

Figure 2 shows a schematic diagram of the DOC+SCRoF+AMOX layout, top part, and the actual physical setup in test cell, bottom part. Whenever possible, exhaust pipes were insulated to reduce heat loss.

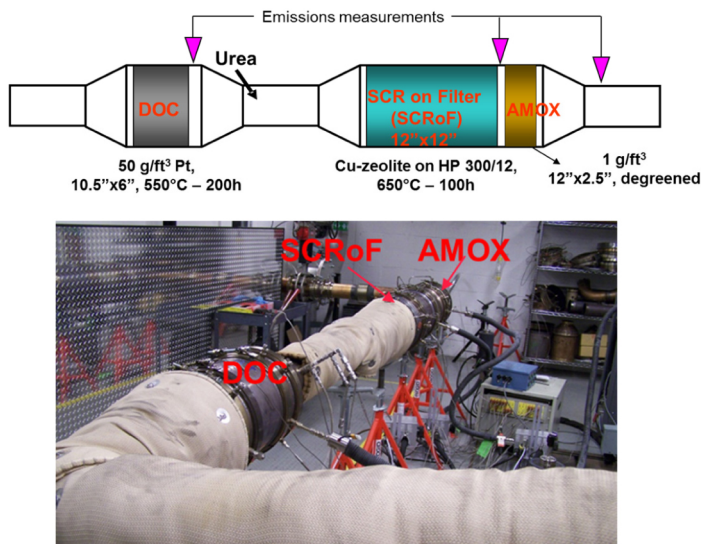


Figure 2. Schematic system layout (top) and test cell exhaust aftertreatment layout (bottom).

Having the capability of simultaneously measuring gas phase compositions is extremely helpful to understand the chemistry occurring in each individual component. During evaluation tests, three MKS FTIR samplers were positioned at DOC outlet, SCRoF outlet, and tail pipe, respectively, for gaseous emissions measurements. Exhaust sampling lines were heated at constant 190°C. Two Horiba direct benches were also positioned at engine-out and at tail pipe, for monitoring THC, CO and O₂ concentrations in the exhaust.

In cases when filter weighing was performed, the part was placed in an oven and held at constant 200°C for 2h before the weight was recorded.

RESULTS AND DISCUSSION

This section shall be divided into three parts. In the first part, SCRoF component and system level NO_x reduction and NH₃ slip characteristics are presented. The second part discusses the DOC+SCRoF+AMOX system sulfation using high sulfur diesel fuel and the subsequent desulfation for activity recovery. In the third part, results from soot loading and passive soot oxidation experiments with both SCRoF and conventional DOC+CSF systems are reviewed. Reaction kinetics modeling was also utilized to explain the experimental observation. Several possible methods to enhance passive soot oxidation are then suggested and discussed.

SCR Performance

Steady-state (SS) engine-bench testing was run to check NO_x reduction across the SCRoF catalyst alone under several engine mode conditions such as: A100, C100 and C20 - for those readers not familiar with load points, the letter is relative speed, A=slow, C=fast; and the number is relative load in %. The target urea dosing ratio was stoichiometric, i.e. NSR=1.0. And filter was regenerated prior to experiment - residual soot in filter should be insignificant.

Results are summarized in Figure 3. At maximum exhaust temperature of 465°C at A100, there was 90% NO_x conversion and SCRoF NH₃ slip was low at 8 ppm. Slightly higher NO_x conversion at 92% was obtained at the maximum exhaust flow rate (C100) and 400°C. At the low temperature condition of 250°C (C20), the DOC generated extremely high NO₂/NO_x ratio at 74%, which is far above the optimal ratio of 50%. In spite of such an unfavorable ratio, the SCRoF part was still able to convert 87% NO_x. Across the temperature spectrum (250-465°C), NH₃ slip out of SCRoF was low. This suggests even higher NO_x conversion efficiency is achievable by slightly over-dosing without exceeding NH₃ slip targets or the use of an AMOX catalyst downstream of the SCRoF.

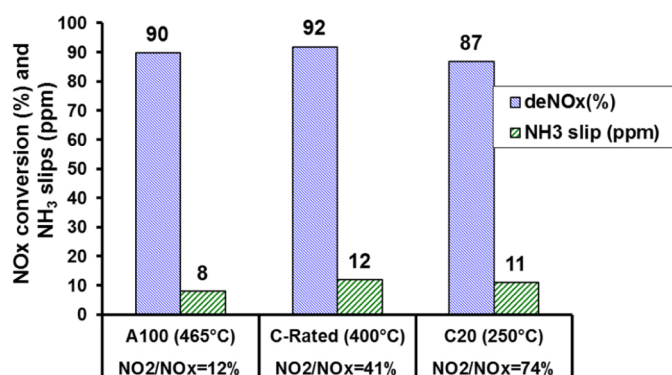


Figure 3. Steady-state SCRoF SCR NO_x conversion and NH₃ slip under three different engine operating modes.

To demonstrate transient cycle performance, a sequence of 1 Cold + 3 Hot NRTC tests (there was a 20 minute soak in between cycles) was run with the DOC+SCRoF+AMOX system. In this test, urea was slightly overdosed at NSR=1.05. Shown in Figure 4, when started with a soot clean filter, the system delivered 92.6% cycle cumulative NO_x conversion efficiency. When the same test started with a 6.2 g/L soot loaded filter, another 3% (abs.) increase of NO_x conversion was obtained. This is presumably due to NO₂/NO_x ratio effect. Over the cycle, the 50 g/ft³ Pt-only DOC generated very high concentrations of NO₂, resulting in a cycle cumulative NO₂/NO_x ratio of 68%. In the highly loaded filter, some of the NO₂ is expected to be consumed as it flows through the soot cake layer. This consumption reduces the NO₂/NO_x ratio closer to the optimal point of 50% before reaching the SCR catalyst in the wall, which helps NO_x reduction.

The transient second-by-second NH₃ concentration was monitored by FTIR at tailpipe after the AMOX brick. Shown in Figure 5 are the temperature and NH₃ slip profiles. In the cold start cycle, after the engine was soaked overnight, urea was not injected due to low temperature until ~5 minutes into the cycle. Consequently, very low NH₃ slip was measured. Once the catalyst reached an equilibrium NH₃ level in the following hot start cycle, there was a small NH₃ spike (of a few ppm) in the first half cycle during the temperature ramp up. The peak NH₃ slip value was fairly low in both soot loading conditions (clean and 6.2 g/L), always under 5 ppm, far below the legislative target of 20 ppm. This demonstrates the system, equipped with an AMOX at the low precious metal loading of 1 g/ft³ Pt, is well positioned to meet future regulations.

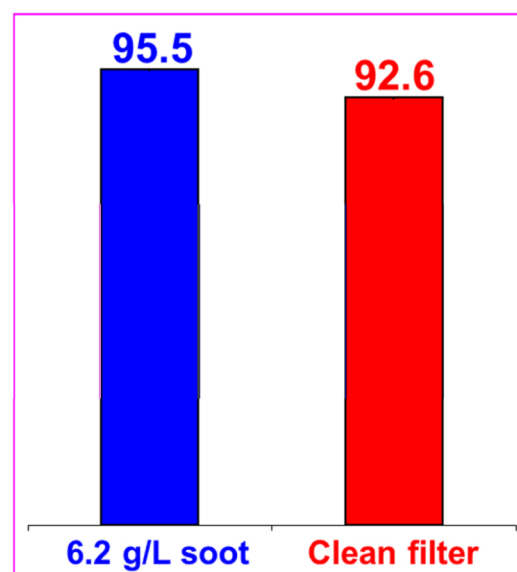


Figure 4. Transient cycle cumulative NO_x conversion efficiency of the tested DOC+SCRoF+AMOX system started with a heavily soot loaded and with a clean filter (reported values are the average of the last three hot cycles). Note SCRoF in NO₂ concentrations were high with a cycle cumulative NO₂/NO_x ratio at ~68%.

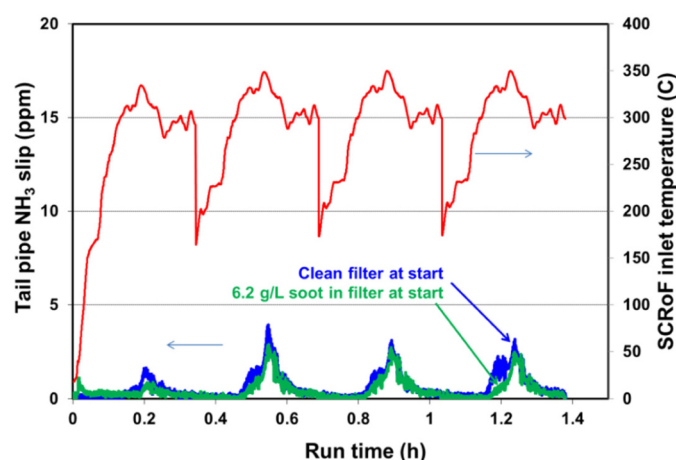


Figure 5. The temperature profile (red line in the right Y-axis) and sec-by-sec tail pipe NH₃ slip profiles (green and blue lines in the left Y-axis) in the 1 cold + 3 Hot transient testing cycle (20mins soak in between cycles).

In summary, a NO_x conversion efficiency greater than 92% was demonstrated during the transient test cycle (NRTC), with a 100h-650°C aged SCRoF. Soot load did not negatively affect NO_x reduction and helped in conditions with excess NO₂. And NH₃ slip was well controlled under both clean and soot-loaded condition.