

Development of SCR on High Porosity Substrates for Heavy Duty and Off-Road Applications

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ABSTRACT

Selective Catalytic Reduction (SCR) catalysts have been demonstrated as an effective solution for controlling NO_x emissions from diesel engines. There is a drive to reduce the overall packaging volume of the aftertreatment system for these applications. In addition, more active SCR catalysts will be needed as the applications become more challenging: e.g. lower temperatures and higher engine out NO_x, for fuel consumption improvements. One approach to meet the challenges of reduced volume and/or higher NO_x reduction is to increase the active site density of the SCR catalyst by coating higher amount of SCR catalyst on high porosity substrates (HPS). This approach could enable the reduction of the overall packaging volume while maintaining similar NO_x conversion as compared to 2010/2013 systems, or improve the NO_x reduction performance for equivalent volume and NH₃ slip. In this work, systems consisting of SCR coated on high porosity substrates were evaluated in comparison to standard substrate based SCR systems used in typical 2010 applications. Performance testing was carried out on laboratory reactor and on engines over various test cycles (Federal Test Protocol (FTP), Ramped Modal Cycle (RMC) and Non-road Transient Cycle (NRTC)). The results indicate improved NO_x conversion with high WCL SCR catalysts on high porosity substrate over standard washcoat loading (WCL) SCR on conventional substrate. In addition, NO_x conversion is improved by increasing cell density from 400 to 750 cpsi. Overall, a volume reduction of 40-50% is achieved by coating SCR on high cell density, high porosity substrate, while maintaining similar NO_x reduction efficiency. The higher amount of SCR catalyst in the HPS allows for higher NH₃ storage and improved NO_x conversion at lower temperature, thus improving performance under transient cycles. This work demonstrates that the higher performance achieved using highly loaded high porosity and high cell density substrates can enable significant volume reduction for future heavy duty and off-road diesel aftertreatment systems.

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1. INTRODUCTION

Diesel engine exhaust is a significant source of air pollution. Strict emission standards have been introduced in several countries to minimize exhaust pollutants. The use of catalytic diesel exhaust aftertreatment systems is one strategy implemented by engine OEMs to meet stringent emission standards, while maintaining peak operating efficiency. Upcoming regulations will be focused on improved fuel efficiency and reduced greenhouse gas emissions (CO_2 , N_2O , CH_4 etc). The various approaches to meet future regulations (e.g. waste heat recovery, hybridization, advanced injection timing...) may result in more challenging conditions for the aftertreatment catalysts: lower operating temperatures and higher engine-out NOx emissions. In addition, there is a drive to reduce the packaging volume of the aftertreatment systems.

Typical exhaust aftertreatment systems may be comprised of a diesel oxidation catalyst (DOC) for the oxidation of HCs and carbon monoxide (and some NO to NO_2 to promote passive filter regeneration and increase the low temperature performance of downstream NOx conversion components), a catalyzed soot filter (CSF) to trap and oxidize PM, a selective catalytic reduction catalyst or a lean NO_x trap (LNT) for NO_x reduction and an ammonia slip catalyst to oxidize selectively the ammonia to N_2 , or combinations thereof [1, 2, 3, 4, 5]. More active SCR catalysts will be required to meet the challenges of lower operating temperatures, higher engine-out NOx emissions and catalyst downsizing.

More active catalysts can be achieved by increasing the number of active sites and increasing the geometrical surface area of the substrate. One approach to increase the active site density is to coat more SCR material on high porosity substrates [6, 7, 8]. The benefit of the HPS is that the SCR washcoat penetrates the substrate wall, thereby allowing more washcoat to be present. In addition, the increased amount of SCR catalyst allows for higher NH_3 storage. These aspects enable improved NOx reduction at equivalent NH_3 slip and catalyst volumes as compared to 2010/2013 systems, or a reduction in packaging volume while maintaining similar NOx conversions and backpressures. by increasing the substrate cell density [9, 10, 11].

The objective of the present study is to compare the performance of SCR catalyst coated on high porosity flow through substrates and standard substrates and determine any potential volume reduction. Cold flow backpressure data will be presented showing the effect of washcoat loading on backpressure. Bench reactor data will be presented showing the effect of temperature, washcoat loading and cell density on NOx reduction performance. Results from steady-state testing, RMC and transient cycles (FTP and NRTC) [12] will be presented demonstrating the NOx conversions and volume reductions that are possible in real systems as compared to standard substrate based SCR systems used in typical 2010/2013 applications.

2. EXPERIMENTAL

Cu-SCR catalyst (Cu-zeolite) was coated on standard substrates at a standard washcoat loading, while the high porosity substrates were coated at a high washcoat loading, with one exception where the 300 cpsi HPS was also coated at standard (std) washcoat loading. The initial washcoat loading on HPS (termed high WCL in section 3.1) was designed to maximize the utilization of the higher void volume in the substrate walls of the HPS.

Bench reactor testing was conducted on 1×1.5 in (25.4×38.1 mm) cores taken from larger parts (5.66×6 in or 10.5×6 in), while engine testing was conducted on 10.5×6 in and 10.5×3.6 in catalysts and combinations thereof. Samples used for bench reactor testing were hydrothermally aged at 650°C for 100 hours in 10% H_2O , while SCR catalysts for engine testing were hydrothermally aged at either 620°C or 650°C for 100 hours, depending on the application.

2.1. Bench Reactor Testing

Bench reactor data were collected using a stainless steel reactor. Data were collected on 1×1.5 in (25.4×38.1 mm) cores at a space velocity of $100\,000\text{ h}^{-1}$. Catalysts for bench reactor testing were hydrothermally aged at 650°C for 100 hours in 10% H_2O . All tests were run with 350 ppm NO, 350 ppm NH_3 , 10% O_2 , 5.0% CO_2 and 4.6% H_2O in the gas feed. The flows of reactant gases and diluent (N_2) were controlled by mass flow controllers. Inlet and outlet components were analyzed using an FTIR bench.

Steady-state conversions were measured at temperatures between 200 and 525°C . The reactor was held at each temperature for an extended period of time (typically around 20 minutes to ensure steady-state was reached. Heated lines were used to deliver gas from the reactor exit to the analyzers.

Transient NH_3 adsorption testing was conducted at 275°C . Approximately 350ppm of NH_3 in N_2 was delivered until steady state was reached and held for an extended period of time (typically 5 minutes). Then the NH_3 was turned off and 350 ppm NOx was turned on. Heated lines (191°C) were used to deliver gas from the reactor exit to the analyzers.

2.2. Engine Testing

Engine tests (steady state, NRTC, FTP and RMC) [12] were run on a 2007, 9L diesel engine to determine the performance of the system. The engine out NOx was approximately 1.5 - 1.6 g/hp-hr. Ultra low sulfur diesel fuel was used during all tests. NH_3 was supplied in the form of an aqueous urea solution injected in front of the SCR catalysts using an air-assisted injection system. A static mixer was placed after the urea injector to maximize reductant mixing.

Emissions were measured using a Horiba Mexa 7400D Series bench at the engine out and SCR out positions and an MKS 2030-HS FTIR at the SCR out position. Heated lines (191°C) were used to deliver the exhaust to the analyzers. The data from the Horiba bench were used to calculate the NO_x conversion across the SCR. FTIR data were used to determine N₂O formation, and NH₃ slip. The catalysts were preconditioned before the beginning of each set of tests to remove any stored NH₃ and soot; the NH₃ concentration at the SCR outlet was measured during this process to verify all the NH₃ had been removed.

2.2.1. Steady-State Tests

3-mode steady-state tests were collected for a DOC + SCR system. The DOC was aged at 700°C / 100 hours, while the SCR catalyst was aged at 620°C for 100 hours. The target ammonia to NO_x ratio (ANR) was 1.05. The inlet temperature of the SCR catalyst was varied from 210-275°C and the space velocity was 107,000 h⁻¹ (based on SCR volume). The NO₂/NO_x in this test was 13% at 275°C.

2.2.2. Cycle Tests

NRTC testing was completed with systems comprised of DOC + SCR. The DOC was aged at 700°C / 100 hours, while the SCR was aged at 620°C for 100 hours. Five repeat NRTC tests were performed. The engine was allowed to cool until the SCR catalyst bed temperature reached 50°C before running the first test. There was a 20 min soak after each of the first two NRTCs; three subsequent tests were run consecutively, without soak. The average ANR during the NRTC was 1.05 - 1.10. The average inlet temperature of the SCR catalyst was for the hot NRTCs was ~317°C and NO₂/NO_x was ~ 20%. Results from the cold, warm and average hot tests will be presented.

Heavy duty transient cycle (FTP) data were collected with a system comprised DOC + CSF + SCR catalysts. In these studies, either two 10.5×6" or one 10.5×6" SCR brick were utilized. The DOC and CSF were aged at 700°C for 100 hours with 10% water and the SCR catalyst was aged at 650°C for 100 hours. Five repeat FTP tests were conducted. The system soaked for 3 hours before the start of the cold FTP and for 20 minutes between cycles. The average ANR during the cycle was 1.00 - 1.10. During the FTP cycle, the inlet average inlet temperature of the SCR catalyst was approximately 235°C. The results from only the hot FTP will be discussed.

The ramped mode cycle (RMC 2010) testing was completed using the same set-up as the FTP. For the RMC, the system soaked for 45 minutes before the start of the test and the cycle was repeated three times with a 45 minute soak between each test. During the cycle, an over and under dosing strategy was used at certain modes; at low speed modes there was over injection to store NH₃ in the new SCR catalyst with high storage capacity and then the urea dosing was reduced at high speed modes to better control NH₃ slip. The ANR used for specific modes will be discussed in the results section. In this

paper, only the last data set of RMC is used. The reason for choosing the last data set of RMC was to make sure the system was stabilized and the data were reproducible.

3. RESULTS AND DISCUSSION

3.1. Comparison of Cu-SCR Performance on Standard and HP Substrates at Different Cell Densities

3.1.1. Cold Flow Pressure Drop

Cu-SCR catalyst coated on standard porosity substrates (300/5 - 750/3) at standard washcoat loading was compared to Cu-SCR catalyst coated on HPS (300/6 - 750/3.5) at high WCL. Additionally, HPS at 300 cpsi was coated at standard loading for a direct comparison to the standard substrate.

Cold flow backpressure data of 5.66×6" bare and coated substrates were collected at 200 cubic feet per minute (Figure 1). The absolute backpressure of the bare HPS (300/6) is slightly higher than the bare standard (300/5) substrates. The higher backpressure is due to the thicker walls of the HPS. The coated standard substrates show a lower backpressure increase compared to the HPS, due to the lower amount of catalyst coated on them. The backpressure increase for the high washcoat loaded HPS was 90 - 110% compared to 50 - 60% for the standard substrates. The absolute backpressure for the 750cpsi is not shown, because the substrate size was not the same as the rest. Comparing the backpressure increase for the standard and HP substrates at the same washcoat loading, it can be seen that the % increase for the HPS is lower (31% vs. 40%), while the absolute backpressure is about the same (2.4 in H₂O). The backpressure contribution from the washcoat was not as significant for the HPS because the coating process was optimized to place a significant amount of the washcoat inside the substrate.

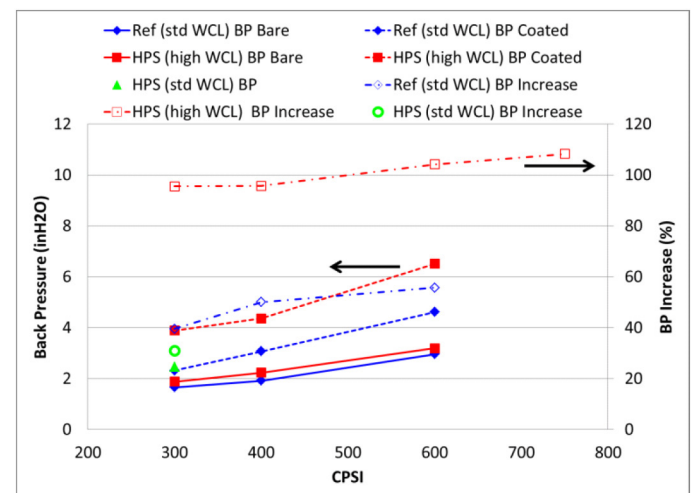


Figure 1. Cold flow backpressure for bare and coated standard (300/5) and HPS (300/6) substrates. The lines with solid symbols correspond to absolute backpressure. The open symbols correspond to backpressure increase.