

Passive Ammonia SCR System for Lean-burn SIDI Engines

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Wei Li, Kevin L. Perry, Kushal Narayanaswamy, Chang H. Kim and Paul Najt
General Motors Global Research and Development

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ABSTRACT

Lean-burn Spark Ignition Direct Injection (SIDI) engines offer potential fuel economy savings, however, lack of cost-effective lean NO_x aftertreatment systems has hindered its broad application. Lean NO_x Trap (LNT) and Urea Selective Catalytic Reduction (SCR) technologies have been widely investigated as possible solutions, but they both have considerable drawbacks. LNT catalysts suffer from high Platinum Group Metals (PGM) cost, poor thermal durability, sulfur poisoning and active SO_x regeneration requirements. Urea SCR systems require a secondary fluid tank with an injection system, resulting in added system cost and complexity. Other concerns for urea SCR include potential freezing of the urea solution and the need for customers to periodically fill the urea reservoir. In this paper we report a low-cost, high efficiency concept that has the potential to be a key enabler for lean-burn gasoline engines. The aftertreatment system includes a close-coupled three-way catalytic converter (TWC) and one or more underfloor SCR catalysts. NH₃ is formed on the TWC during short periods of rich engine operations and the generated NH₃ is then stored on the underfloor SCR catalysts. During the subsequent lean operations, the NO_x that breaks through the TWC converter is converted by the NH₃ stored on the SCR catalysts. Test results during the New European Driving Cycles (NEDC) showed that very high (>85%) lean NO_x conversion efficiencies were achieved with aged converters. The potential and remaining issues of this new concept will be discussed. HC emission reduction is challenging due to the low exhaust temperatures. Further improvements in SCR catalyst technologies are required for high speed (≥100 km/h) lean operations. In summary, passive NH₃ SCR has been demonstrated as an efficient and low cost lean NO_x aftertreatment technology for stratified gasoline engines.

INTRODUCTION

Lean burn spark-ignition, direct injection (SIDI) engines offer significant potential for improving engine efficiency and reducing greenhouse gas emissions [1,2,3,4]. However, cost-effective lean NO_x aftertreatment systems remain a significant technical challenge. LNT and urea SCR technologies have been widely investigated as possible solutions, but both have considerable drawbacks. LNT catalysts suffer from high PGM costs and severe sulfur poisoning, and require periodical, active sulfur regenerations [6, 7]. Recently active urea SCR technology has been proposed for lean gasoline engines and it was suggested that the latest SCR catalyst technologies were thermally durable for such applications [8]. However, urea SCR systems require a secondary fluid tank with an injection system, resulting in added system cost and complexity. Other concerns for urea SCR include the potential freezing of urea solution and the need for customers to periodically fill the urea solution [9]. In this paper we report a low-cost concept of lean NO_x aftertreatment that has the potential to be a key enabler for lean-burn gasoline engines.

For the GM stratified lean SIDI program, we initially focused on the LNT/SCR system for lean NO_x conversion. An SCR catalyst was added downstream of the LNT catalyst aimed to utilize the abundant NH₃ formed during the LNT regenerations for additional NO_x conversion. However, it was also found that during the LNT regenerations there was significant NH₃ formation on the close-coupled three-way catalyst (TWC) from engine-out NO_x. This has led us to probe the concept of passive NH₃ SCR system (Figure 1).

The basic concept includes the following steps:

1. Periodically the engine operates slightly rich of stoichiometry to produce NH_3 on the three-way catalysts from engine out NO_x ;
2. The produced NH_3 is stored on the downstream SCR catalyst;
3. During subsequent lean operations the NO_x emissions after the TWC are reduced on the SCR catalyst by the stored NH_3 ;
4. A multiple SCR configuration may be used to expand the thermal operating window (more details on this in later sections).

<figure 1 here>

The key difference between this concept and other LNT-related technologies is that in this concept there is no need for NO_x storage; rather NH_3 is the component being stored on the SCR catalysts, thereby avoiding the use of expensive LNT catalysts.

In this work we aim to clarify aspects of this concept by evaluating the following issues:

1. Ability of the three-way catalyst to make NH_3 efficiently
2. Ability to store NH_3 on the SCR catalysts and convert NO_x during the subsequent lean operations
3. SCR catalyst durability required for gasoline applications including under redox conditions
4. Aftertreatment system architecture and thermal management

EXPERIMENTAL

A 2.2L stratified-charge direct-injection engine was used for this work and the tests were carried out on a transient dynamometer capable of running simulated NEDC driving cycles. Standard reformulated gasoline fuel with 30 ppm sulfur was used for catalyst aging and testing during this work. An in-house d-SPACE controller equipped with a Micro-Autobox was used for engine control with full authority. Horiba analyzers and MKS FT-IR analyzers were used for measuring exhaust gas compositions at various locations (engine-out, post-TWC, post-SCR, etc.).

The catalysts used in this work include TWC (600/4 substrate, 1 liter) and SCR (400/4 substrate, 2 liters each for SCR1 and SCR2) parts from various catalyst suppliers and were aged on engine dynamometers using GM proprietary accelerated aging protocols. The close-coupled three-way catalysts were aged 50 hours using an aging protocol with the maximum catalyst bed temperature of approximately 1030°C; whereas the SCR catalysts were aged 50 hours with a

maximum catalyst bed temperature of about 760°C. The aging treatments are representative of vehicle aging of 100,000 km. The TWC formulations are similar to production parts and the PGM loadings range from 70 g/ft³ to 200 g/ft³. Both Cu and Fe-based SCR catalysts were evaluated in this work; however, all the data shown in this paper were acquired with aged Cu SCR catalysts.

RESULTS AND DISCUSSION

NH_3 FORMATION ON THREE-WAY CATALYSTS

It has been reported that NH_3 can be formed on the three-way catalysts (TWC) under rich conditions [10]. However, as there may be a fuel penalty for running the engine rich to produce NH_3 , it is important to optimize the engine conditions to maximize the NH_3 formation efficiency and minimize the fuel penalty.

First we carried out air-fuel ratio (AFR) sweep tests to probe the optimum conditions for maximum NH_3 formation. As expected, there was little NH_3 formed when the engine was running at an AFR lean of stoichiometry. The amount of formed NH_3 increased as the AFR ratio was decreased (richer), until it reached a maximum around the AFR of 14.0-14.2. With further decreases in AFR (richer) the amount of formed NH_3 decreased. The reason for this trend is that both NO_x and reductants (H_2/CO) are required in order to form NH_3 ; therefore the formation of NH_3 could be limited by either component. While it is clear that for AFRs lower than 14.2 (richer) the formation of NH_3 is limited by the engine-out NO_x concentrations (the discrepancy between post-TWC NH_3 and engine-out NO_x is likely due to measurement uncertainties), for AFRs higher than 14.2 (leaner) the formation of NH_3 is likely limited by the engine-out reductants (CO/H_2) concentrations. Based on these measurement results, the AFR of 14.2 was chosen as the condition for NH_3 production during the transient cycle operations.

In order to maximize the fuel saving potential, it is critical that the engine operates in the lean combustion mode as much as possible during the NEDC driving cycle. Vehicles with lean burn SIDI engines and LNT catalysts typically operate the engine in stoichiometric AFR for 150-200 seconds after cold starts [4, 5], as this helps to heat up the LNT to its operating temperatures (250°C). In this work the engine was allowed to shift into lean operations as quickly as possible after cold start. Therefore after the engine started, it operated in a catalyst heating mode during which the close-coupled converter was heated to its operation temperature for efficient removal of HC and CO (Figure 3). Afterwards the engine was switched to lean operations for most of the driving cycle.

except certain brief periods during the accelerations. In some of these acceleration cases the engine may be scheduled for stoichiometric operations due to the load requirements. In order to produce NH_3 more efficiently the engine was operated at AFR of 14.2 instead of stoichiometric AFR and many times also preceded by a brief rich tip-in to purge the oxygen in the oxygen storage materials in the TWC.

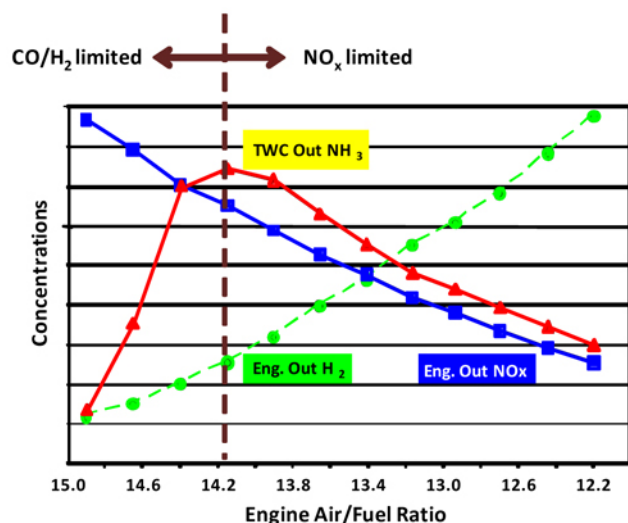


Figure 2. AFR sweep tests for NH_3 formation; 2000 rpm/ 2 bar BMEP; TWC bed temperatures between 550°C to 600°C

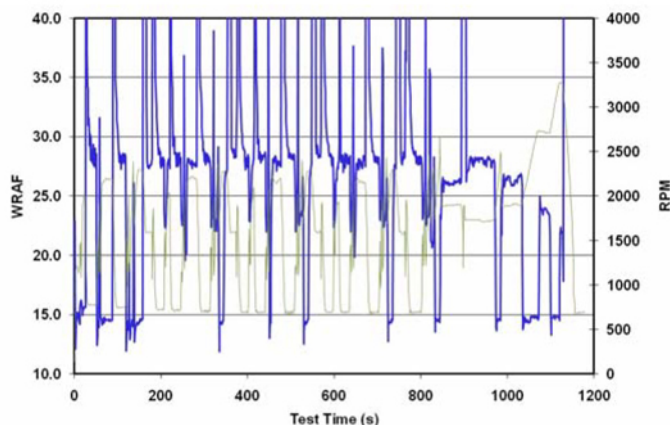


Figure 3. Engine AFRs during an NEDC driving cycle; WRAF: wide range air-fuel sensor signal

During the transient NEDC cycle the rich operations (AFR = 14–14.2) during the accelerations did lead to significant NH_3 formation (Figure 4) with very high conversion efficiencies, confirming the steady state AFR sweep test results. The amount of NH_3 formed and the corresponding fuel penalty are functions of engine control strategy and catalyst compositions. We tested various combinations of the

engine control methods and catalyst compositions and found that it is possible to efficiently produce the required amount of NH_3 with minimum fuel penalty (within test variability, <1%). This conclusion is only based on engine dynamometer test results and the actual case on a vehicle may be different.

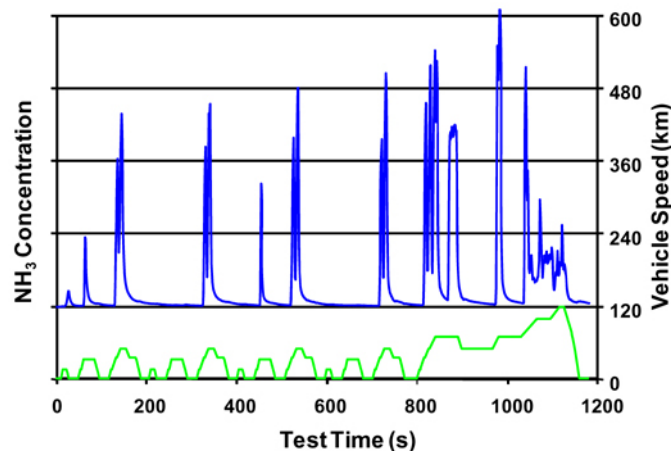


Figure 4. Post-TWC NH_3 concentration profile during the NEDC cycles; blue line: NH_3 concentrations and green line: simulated vehicle speed

NH_3 STORAGE ON SCR CATALYSTS

After NH_3 is formed on the TWC, it must be stored on the SCR catalyst for subsequent lean NO_x conversion. Therefore the NH_3 storage characteristics on SCR catalysts have a significant impact on the performance of the passive NH_3 SCR concept. Two types of zeolite-based catalysts, Cu- and Fe-zeolite catalysts, are commonly used for NH_3 SCR reactions. Typically the NH_3 storage capacity decreases with increasing temperatures and Cu-zeolite catalysts usually have higher NH_3 storage capacity than Fe-zeolite catalysts (Figure 5). NH_3 storage capacity of known catalysts is negligible beyond 400°C, therefore in order to achieve high NO_x efficiencies, the SCR bed temperature must be kept below 400°C.

For a practical catalyst technology, it is critical that it has a broad operating temperature window so that it remains highly efficient under a variety of driving conditions. A comparison of typical thermal operating windows for various lean NO_x technologies with aged catalysts is shown in Figure 6. While LNT catalysts are most effective at temperatures between 250–450°C, the active urea SCR has a broader temperature window as it becomes active at about 180°C and remains effective until 550–600°C. For the passive NH_3 SCR, efficient NO_x conversion relies on the NH_3 stored on the SCR catalyst. Therefore the low temperature limit is similar to the active urea SCR (180°C) as long as there is NH_3 stored