

Ammonia Generation and Utilization in a Passive SCR (TWC+SCR) System on Lean Gasoline Engine

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ABSTRACT

Lean gasoline engines offer greater fuel economy than the common stoichiometric gasoline engine, but the current three way catalyst (TWC) on stoichiometric engines is unable to control nitrogen oxide (NO_x) emissions in oxidizing exhaust. For these lean gasoline engines, lean NO_x emission control is required to meet existing Tier 2 and upcoming Tier 3 emission regulations set by the U.S. Environmental Protection Agency (EPA). While urea-based selective catalytic reduction (SCR) has proven effective in controlling NO_x from diesel engines, the urea storage and delivery components can add significant size and cost. As such, onboard NH_3 production via a passive SCR approach is of interest. In a passive SCR system, NH_3 is generated over a close-coupled TWC during periodic slightly rich engine operation and subsequently stored on an underfloor SCR catalyst. Upon switching to lean operation, NO_x passes through the TWC and is reduced by the stored NH_3 on the SCR catalyst. In this work, a passive SCR system was evaluated on a 2.0-liter BMW lean burn gasoline direct injection engine to assess NH_3 generation over a Pd-only TWC and utilization over a Cu-based SCR catalyst. System NO_x reduction efficiency and fuel efficiency improvement compared to stoichiometric engine operation were measured. A feedback control strategy based on cumulative NH_3 produced by the TWC during rich operation and NO_x emissions during lean operation was implemented on the engine to control lean/rich cycle timing. At an SCR average inlet temperature of 350 °C, an $\text{NH}_3:\text{NO}_x$ ratio of 1.15:1 (achieved through longer rich cycle timing) resulted in 99.7 % NO_x conversion. Increasing NH_3 generation further resulted in even higher NO_x conversion; however, tailpipe NH_3 emissions resulted. At higher underfloor temperatures, NH_3 oxidation over the SCR limited NH_3 availability for NO_x reduction. At the engine conditions studied, greater than 99 % NO_x conversion was achieved with passive SCR while delivering fuel efficiency benefits ranging between 6-11 % compared with stoichiometric operation.

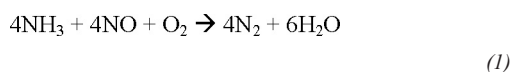
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INTRODUCTION

Concerns about greenhouse gas emissions and dependence on petroleum have motivated governments around the world to develop regulations that require significant improvements in vehicle fuel economy. In the U.S., the fuel economy regulations for light duty vehicles will require the corporate average fuel economy (CAFE) to increase to 54.5 miles per gallon by 2025, which is more than 50 % higher than the current regulations. These regulations have motivated vehicle manufacturers to pursue a range of technologies that reduce petroleum consumption. To ensure their adoption in the marketplace, these technologies must comply with increasingly stringent exhaust emissions regulations for pollutants such as nitrogen oxides (NO_x), carbon monoxide (CO), hydrocarbons (HCs), and particulate matter (PM). The recently approved U. S. Environmental Protection Agency Tier 3 emissions regulations require more than 80 % reduction in NO_x and non-methane HCs from the current Tier 2 Bin 5 emission standard. Meeting both the 2025 CAFE standards for fuel economy and the Tier 3 emissions regulations will require significant advancements in engines, powertrains, and emissions control.

In the U.S., the light duty vehicle fleet is dominated by gasoline engines operating at stoichiometric air-to-fuel ratios (AFR) [1]. Stoichiometric exhaust conditions are suitable for a three way catalyst (TWC) to effectively control NO_x , CO, and HC emissions to regulated levels. Lean operation can increase the fuel economy of a gasoline engine by 10-20 % over stoichiometric operation [2], [3]. While TWCs can effectively oxidize CO and HCs under lean conditions, they are unable to reduce NO_x in the presence of excess oxygen; thus, a different emissions control technology is required for the abatement of NO_x emissions from lean burn gasoline engines. While urea-based selective catalytic reduction (SCR) has proven effective in controlling NO_x emission from diesel vehicles, the urea storage and delivery components can be large and add significant cost. As such, onboard NH_3 production via a passive SCR approach is of interest [4], [5]. In a passive SCR system, NH_3 is generated over a close-coupled TWC during periodic slightly rich operation and subsequently stored on an underfloor SCR catalyst. Upon switching to lean, NO_x passes through the TWC and is reduced by the stored NH_3 on the SCR catalyst. This approach is particularly attractive for lean gasoline engines because it makes use of a TWC that is already onboard and eliminates or greatly reduces the need for a urea system.

The effectiveness of a passive SCR system depends on efficient and selective NH_3 generation and utilization over the TWC+SCR catalysts. Since the rich operation necessary for NH_3 generation incurs a fuel consumption penalty, minimizing the duration and magnitude of rich excursions is critical and requires a combination of engine management and emissions control optimization. During rich/lean cycling, NH_3 produced while rich and NO_x concentration when lean ultimately control cycle timing and fuel consumption [6], [7]. For the downstream SCR, a molar ratio of NH_3 to NO_x of 1 offers the best theoretical NO_x reduction performance with minimal NH_3 emissions due to the stoichiometry of the standard SCR reaction between NH_3 and NO_x as shown in Equation 1.



Of course, achieving this ratio in a passive SCR system may be challenging; for instance, NH_3 lost to oxidation over a SCR poses an additional constraint on system efficiency. This study aims to understand how NH_3 : NO_x ratio and cycle timing affect NO_x reduction efficiency and fuel consumption of a passive SCR system.

EXPERIMENTAL SETUP

Engine Platform

All experiments were conducted on a lean gasoline engine coupled to a motoring direct current dynamometer that controls the engine speed and load. The engine was removed from a European Model Year (MY) 2008 BMW 1-series 120i (E87) vehicle. It is a 4-cylinder 2.0-liter naturally aspirated direct injection gasoline engine that has a rated power of 125 kW at 6700 revolutions per minute (rpm) and a torque of 210 Nm at 4250 rpm. Table 1 summarizes engine specifications.

Table 1. BMW 2.0-liter lean direct injection engine specifications.

Engine Model Number	N43B20
Displaced volume	1995 cm ³
Number of cylinders	4
Stroke	90 mm
Bore	84 mm
Compression ratio	12.0:1
Rated Power	125 kW at 6700 rpm
Rated Torque	210 Nm at 4250 rpm

The engine was operated on an EEE-Lube Certification Gasoline (Haltermann Solutions) with low sulfur levels. Table 2 shows key gasoline fuel properties provided by the vendor.

The engine uses a spray guided combustion system design. In this design, a piezoelectric injector is located at the top center of the combustion chamber, with a spark plug in close proximity to the injector. This geometry allows ignition of the fuel as it is injected, which results in shorter mixture formation time compared to wall guided and air guided combustion systems [8]. With shorter mixture formation time, lean engine operation can be extended to higher

speeds and loads, resulting in additional fuel savings. The overall air-fuel equivalence ratio, or λ (ratio of actual AFR to stoichiometric AFR), during lean operation ranges between 1.3 and 2.2. Up to 20 % reduction in fuel consumption can be achieved with lean operation relative to stoichiometric. The engine, however, only operates lean over a portion of the engine's speed and load range (up to 4500 rpm and 75 % load). Outside the lean operating range, at higher engine speeds (>4500 rpm) and loads (>75 %), the engine operates in the stoichiometric combustion mode. The engine can also operate in the stoichiometric mode over its entire operating range. For this study modifications to the stoichiometric mode were made in order to operate the engine rich ($\lambda < 1$) for NH_3 generation. Details on an additional rich combustion strategy available on this engine can be found in a previous publication [6].

Table 2. EEE-Lube Cert Gasoline fuel properties.

Lower Heating Value, kJ/kg	42715
Density, kg/liter	0.742
C, weight fraction	0.8598
H, weight fraction	0.1314
Oxygen, weight %	<0.01
Sulfur, mg/kg	4
Aromatics, volume %	27.4
Olefins, volume %	0.9
Saturates, volume %	71.8
Research Octane Number (RON)	96.4
Motor Octane Number (MON)	88.4

The factory engine control unit was replaced with a custom full-pass control system developed by National Instruments - Powertrain Controls Group. The controller mimics the original equipment manufacturer (OEM) combustion strategies and, furthermore, enables full control of all engine parameters, including fuel injection quantity and timing, spark timing, λ , and EGR rate, which allows full control of the different modes of combustion. Additional information on the engine setup, performance and emissions can be found in [2], [3], [6].

Exhaust System

The exhaust system consisted of a 1.3 L closed-coupled TWC and a 2.5 L Cu zeolite SCR installed downstream of the TWC. The location of the SCR catalyst is representative of an underfloor position, which is best suited to the temperature range for optimal NO_x reduction performance by the SCR catalyst. The engine and the exhaust configuration are shown in Figure 1. The TWC is a Pd-only formulation with 7.33 g/L of Pd. This catalyst is the front half of a commercial dual zone TWC used in a 2009 PZEV (Partial Zero Emissions Vehicle) Chevrolet Malibu. For this study, the front portions of three Chevrolet Malibu TWCs were cut out, mounted in series in a stainless steel can and installed in the engine exhaust to provide adequate catalyst volume for the experiments. The SCR catalyst was provided by Umicore. It is a small pore Cu-zeolite catalyst on a 400 cpsi substrate. Both of the catalysts were degreened in the engine exhaust for approximately 20 hours with maximum inlet temperatures of 850 °C and 700 °C for the TWC and SCR respectively.

Emission Sampling

Raw exhaust was sampled at the engine out, post TWC and post SCR locations. The sample was routed through a heated filter prior to analysis. A California Analytical Instruments (CAI) heated flame ionization detector (FID) was used to measure total unburned hydrocarbons. An MKS Instruments MultiGas Model 2030 HS FTIR Spectrometer with a heated 5.11 m multipass gas cell was utilized to measure multiple gases, including CO, CO₂, nitrogen monoxide (NO), nitrogen dioxide (NO₂), nitrous oxide (N₂O) and ammonia (NH₃).

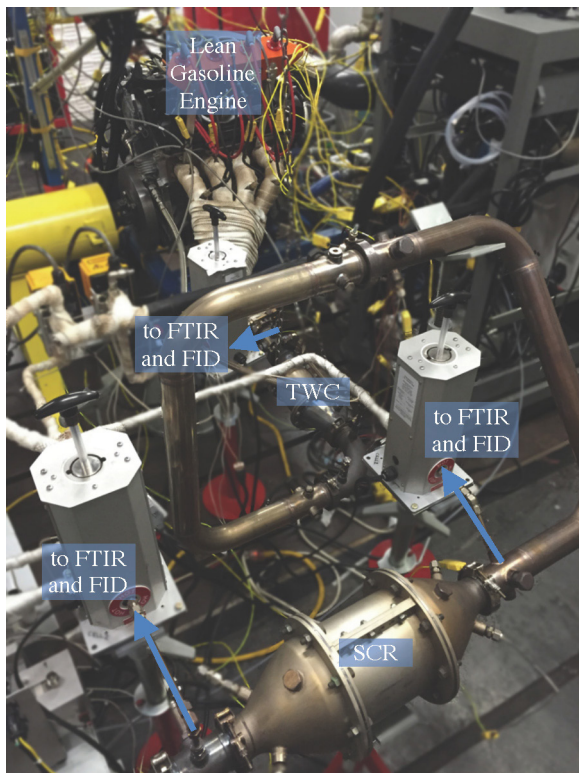


Figure 1. Lean gasoline engine with a close-coupled TWC and an underfloor SCR.

Experimental Conditions

To assess NO_x reduction efficiency of a passive SCR system and fuel efficiency improvements under realistic operating conditions, a series of rich/lean cycling experiments was performed where the engine alternated between rich and lean operation. These experiments were designed to be representative of the conditions a passive SCR system would encounter under realistic driving conditions. Acceleration events create an opportunity for higher engine out NO_x and, thereby TWC NH₃ production, which can result in additional fuel efficiency gains in a passive SCR system [7]. In this study, load step cycling experiments were carried out to simulate acceleration driving events. During load step experiments, the engine alternated between rich operation ($\lambda=0.97$) at 8 bar brake mean effective pressure (BMEP) and lean operation at 2 bar BMEP while maintaining 2000 rpm engine speed. A feedback control strategy based on cumulative NH₃ produced by the TWC during rich operation and NO_x emissions during lean operation was implemented with the engine controller. In this manner, control setpoints for cumulative NH₃ production and

NH₃:NO_x ratio allowed control of the lean/rich cycle timing to achieve the desired NH₃:NO_x ratio. The rich cycle timing during the experiments with NH₃:NO_x=1, which served as a starting point, was such that the NH₃ produced during rich operation would use approximately 25 % of the NH₃ storage capacity of the SCR catalyst based on prior flow reactor measurements. The NH₃ capacity utilization was intentionally kept low to prevent NH₃ slip during the lean phase, which was observed in flow reactor experiments with higher NH₃ loadings.

RESULTS AND DISCUSSION

Effects of NH₃:NO_x

For the SCR NO_x reduction, a molar ratio of NH₃ to NO_x of 1 represents a theoretical minimum NH₃ production requirement for complete NO_x reduction due to the stoichiometry of the reaction between NH₃ and NO_x. Since, in practice, some of the NH₃ stored on the SCR catalyst will oxidize, more NH₃ must be produced to account for NH₃ lost to oxidation. Figure 2 shows the overall NO_x reduction efficiency of the passive SCR system during load step lean/rich cycling experiments. The cycle average temperature at the SCR inlet during these experiments was 350 °C. As seen in Figure 2, at the theoretically optimal NH₃:NO_x ratio of 1, 95.1% NO_x reduction was achieved. Increasing NH₃:NO_x resulted in increasing NO_x conversion, achieving 99.5 % NO_x conversion with 13% excess NH₃. Increasing NH₃:NO_x further only resulted in incremental NO_x reduction benefit. At this temperature, NH₃ can be readily oxidized by oxygen present in the exhaust, so extra NH₃ had to be produced to increase NO_x reduction efficiency [9], [10].

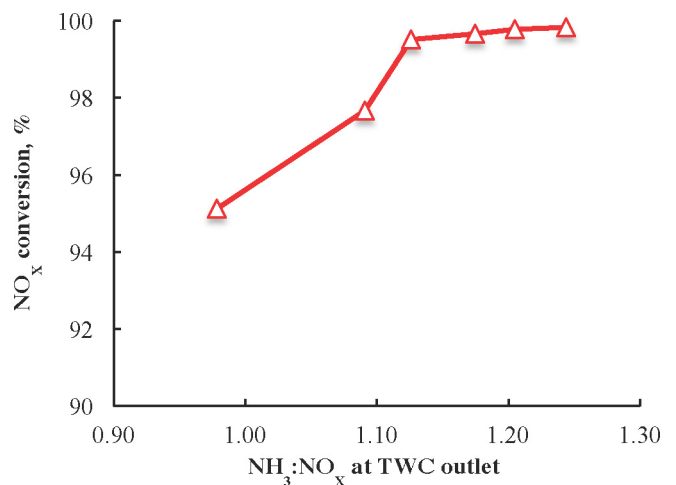


Figure 2. Overall NO_x conversion as a function of NH₃:NO_x ratio during load step rich/lean cycling experiments. Rich conditions: 2000 rpm, 8 bar BMEP, and $\lambda=0.97$. Lean conditions: 2000 rpm, 2 bar BMEP, and $\lambda=2.0$. Average temperature at TWC inlet=470 °C and SCR inlet=350 °C

The N-species product distribution at the tailpipe is shown in Figure 3. Figure 3a shows the cycle averaged N-species emissions as a function of NH₃:NO_x, and Figure 3b shows example N-species concentration as a function of time over two lean-rich cycles to aid in visualizing the cycle-based behavior. Increasing NH₃:NO_x decreases