

Development and Demonstration of LNT+SCR System for Passenger Car Diesel Applications

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ABSTRACT

The regulations for mobile applications will become stricter in Euro 6 and further emission levels and require the use of active aftertreatment methods for NO_x and particulate matter. SCR and LNT have been both used commercially for mobile NO_x removal. An alternative system is based on the combination of these two technologies. Developments of catalysts and whole systems as well as final vehicle demonstrations are discussed in this study.

The small and full-size catalyst development experiments resulted in PtRh/LNT with optimized noble metal loadings and Cu-SCR catalyst having a high durability and ammonia adsorption capacity. For this study, an aftertreatment system consisting of LNT plus exhaust bypass, passive SCR and engine independent reductant supply by on-board exhaust fuel reforming was developed and investigated. The concept definition considers NO_x conversion, CO₂ drawback and system complexity. The passive SCR significantly contributes to the total NO_x conversion over a broad temperature range. In contrary to conventional LNT applications, the LNT NO_x slip does not directly reduce the system NO_x conversion but can be converted over the passive SCR. Thereby, the NO_x adsorption duration might be prolonged, resulting in lower LNT regeneration frequency and less fuel consumption, resp. CO₂ emissions. Additionally, the engine independent LNT enrichment is beneficial regarding fuel consumption in comparison to conventional engine internal enrichment.

The system has been tested in a vehicle in combination with a highly efficient Diesel engine. Results regarding CO₂ and NO_x as well as their dependency on temperatures are shown here.

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INTRODUCTION

The upcoming worldwide more stringent emission legislation and CO₂ restriction are highly challenging for modern Diesel engine and aftertreatment technology. Beside the more restrictive emission limits, new emission evaluation test procedures, like WLTP (Worldwide harmonized Light vehicles Test Procedure), RDE (Real Driving Emissions) and PEMS (Portable Emissions Measurement System) result in enlarged emission relevant engine operation area. Beside the increased

coverage of engine load and speed also the emission testing including cold start and high temperature cycles demands an extended active temperature range of aftertreatment systems. On the one hand, moving to lower absolute emission levels by engine internal measures progressively cause drawbacks in CO₂ emission and system cost and complexity. On the other hand, increased combustion efficiency and decreased friction losses by improved engine technologies lead to lowered exhaust gas temperatures and raise the kinetic based

challenges of exhaust aftertreatment functionalities. For meeting all emission requirements with respect to the typical trade-off of pollutant minimization and CO₂ emission, an optimum interaction of advanced engine internal and exhaust aftertreatment functionalities is demanded.

On the aftertreatment side, advanced combined systems offer the best overall performance by the benefit of synergistic effects. Beside the well-established DPF-technology, the combination of the two already commercially applied NO_x removal technologies LNT (Lean NO_x Trap) and SCR (Selective Catalytic Reduction) is a suitable approach due to the specific single characteristics [1,2,3]. The LNT offers low temperature activity by intermediate NO_x adsorption, but is thermodynamically limited at high temperature. The urea based SCR offers high temperature activity, but is limited at cold conditions due to dosing release of liquid urea as well as high dependence on NO₂/NO_x ratio. In general, LNT and SCR combination systems can be classified in 3 types:

1. LNT based system: LNT + passive SCR (operated without urea dosing system)
2. SCR based system: passive NO_x adsorber (PNA) + active SCR (operated with urea dosing)
3. LNT + SCR based system: LNT + active SCR (operated with urea dosing)

In an LNT based system, the SCR contributes passively to the total NO_x conversion by utilization of the LNT's intrinsic ammonia (NH₃) formation during rich regeneration without the disadvantages of a urea dosing system, such as additional hardware costs or urea refilling efforts. However, the NO_x conversion performance is still mainly dominated by the LNT behavior, i.e. by NO_x adsorption capacity and NH₃ formation. The intended task of a PNA in an SCR based system is intermediate NO_x storage during cold start and NO_x release after SCR light-off. The SCR is conventionally operated by active urea dosing control. The highest NO_x conversion potential is given by combination of two active systems, i.e. LNT including rich regeneration and urea based SCR. However, the LNT+SCR based system shows maximum packaging, costs and system complexity issues.

In this study, an LNT based system was developed and investigated as concept study. New ECU software functionalities for LNT+SCR control have been developed on the level of rapid control prototyping (RCP). Future sensor layout strategies have not been investigated. The concept is in line with the requirement specification of the 7th Framework Program of the European Union, demonstrating emissions of 10% below EU6 legislation (e.g. NO_x emission of 72 mg/km) without urea dosing system at CO₂ emission of 98 g/km. Additionally, the system performance needs to be demonstrated in real driving conditions (RDE), too. To fulfill the

requirements especially with strong focus on CO₂ emission, the concept is based on engine independent LNT regeneration by integrated on-board exhaust fuel reforming. As discussed in [4], fuel reforming showed much higher reductant generation efficiency than conventional Diesel engine internal enrichment. The investigated aftertreatment system is described in more detail in the next chapter.

Aftertreatment Concept Description

The investigated LNT based aftertreatment system consists of LNT plus exhaust bypass, passive SCR and on-board exhaust gas fuel reforming. The final concept layout and the simplified method of operation are shown schematically in figure 1.

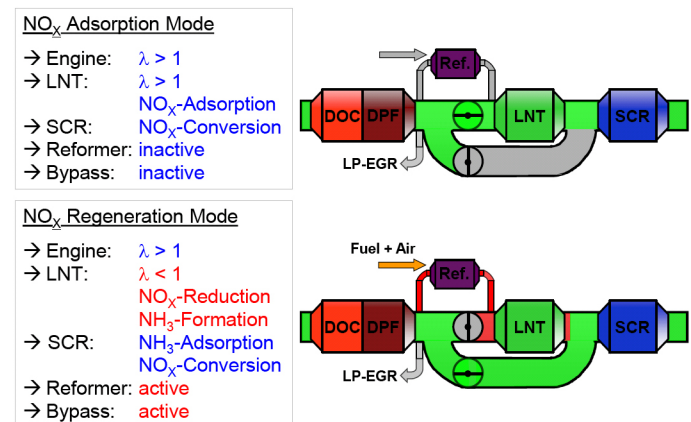


Figure 1. Simplified method of operation of final layout of LNT based combination system, with under-floor LNT plus exhaust bypass, passive SCR and integrated exhaust gas fuel reformer.

Basically, the system is discontinuously operated in two operation modes. During NO_x adsorption mode, the total system is operated under lean condition. The LNT path is opened, the LNT bypass is closed. NO_x treatment takes place either by NO_x adsorption on the LNT or continuous reduction on the SCR by previously stored NH₃. The integrated on-board fuel reformer is inactive. For the regularly necessary LNT regeneration, the LNT is locally enriched by opening the bypass path, closing the LNT path and feeding H₂ and CO containing reformat gas at low space velocity via the activated fuel reformer. The exhaust paths are actuated by two serial exhaust flaps. The on-board exhaust fuel reformer is activated on demand and is predominantly operated by exhaust gas, which is taken downstream DPF. Diesel is injected by an air-assisted, low-pressure injector at a significant under-stoichiometric air/fuel ratio ($\lambda_{\text{Ref}} = 0.35-0.80$) and is catalyzed to a H₂- and CO-rich synthesis gas. The engine is continuously operated at optimum efficient lean A/F ratio. Conventionally, LNT regeneration is realized by engine enrichment. This non-typical Diesel operation is usually causing drawbacks, e.g. highly decreased engine internal combustion efficiency, oil dilution and drivability and noise issues. Beside the target

product N_2 , NH_3 is intrinsically formed and emitted as by-product. Conventional LNT applications usually need to avoid this NH_3 emission during the rich LNT regeneration. In contrast, NH_3 is stored on the downstream positioned SCR in the combination system. The continuously consumed SCR NH_3 load during the NO_x adsorption mode is getting refilled only during LNT regeneration. Thus, the SCR is operated discontinuously. The lean tailpipe exhaust gas composition offers the possibility to oxidize the reductant slip during LNT regeneration.

Beside the possibility for engine independent LNT enrichment at low LNT space velocity, the bypass concept offers further advantages regarding thermal and chemical ageing issues. By opening the LNT bypass during regular DPF regeneration or irregular DPF thermal shock, the LNT can be protected from critical high exhaust gas temperatures. Additionally, by placing a sulphur storage function upstream the LNT on DOC or DPF, the sulphur exposure of the LNT can be minimized. Desulphation can be done over open LNT bypass. Hence, high thermal degradation by desulphation can be minimized. In case of an LNT desulphation, the H_2S emission from the LNT can be continuously oxidized on or downstream of the permanently lean operated SCR catalyst. The LNT desulphation will be done in parallel during DPF regeneration by reformat gas dosing. Gas dosing into the open LNT path and oxidation on LNT result in engine independent LNT heat up. By switching the LNT valve, conventional lean/rich wobbling is applied.

EXPERIMENTAL

Catalyst Preparation

The LNT and SCR catalysts for small- and full-scale experiments were based on metallic substrates (500/600 cpsi, foil thickness 50 μm), where a porous coating with active metals and adsorbents were added (details by [5]). Thermally stable rare earth element (like ceria) based oxygen storage and thermal stabilizer material were added as promoters for alumina based LNTs having platinum (Pt) and rhodium (Rh) as active metals. Two different LNT formulations ("LNT A" and "LNT B") were tested, which consisted of different sets of NO_x adsorbents and OSC was also slightly different. The surface area (BET) of LNT coatings was between about 130-140 m^2/g . The volume of LNT converter was 1.4 dm^3 , the Pt loading was 80 ("PGM low") or 130 g/cft ("PGM high") and the Rh loading was 5 g/cft in engine and vehicle experiments. SCR catalysts had copper (Cu) and iron (Fe) as active metals. The coating amount was about 140 g/L. The surface area (BET) of Cu and Fe catalysts was about 400-500 m^2/g . The volume of the SCR converter was 1.95 or 2.9 dm^3 in engine and vehicle experiments.

Laboratory Testing with Simulation Gases

The activity of small-size samples were evaluated in laboratory experiments, where a cycle includes longer lean and short rich periods simulating LNT and LNT + passive SCR system conditions (experimental details by [5]). The catalyst samples (diameter 14 mm) were located in quartz reactors heated by an IR furnace. The lean-rich cycles were repeated for five times with the mixtures presented in [table 1](#). The conditions in by-pass system were simulated with an air addition between LNT and SCR units.

Table 1. Experimental conditions in laboratory simulations.

Mixture	Lean	RichA	RichA+air
NO_x , ppm	500	1500	1065
C_3H_6 , %	0.1	0.1	0.07
CO , %	0.25	5.73	4.07
H_2 , %	0.08	2.0	1.42
CO_2 , %	10	10	7.1
H_2O , %	10	10	7.1
N_2	bal.	bal.	bal.
λ	1.41	0.81	1.20
Period, s	30-120	5-15	5

Engine-Bench Experiments for Catalyst Development

The engine experiments were conducted with 1.2L R3 turbocharged Diesel research engine equipped with DOC+DPF system before LNT+SCR units. The experimental conditions were simplified for catalyst development purposes not to use efforts to manage with the whole EATS with by-pass and reformer gas during the catalyst evaluations. Therefore, these experiments were done using all-in-line system together with the engine management system for periodic lean-rich operation. Lean-rich driving is always transient conditions, but the experimental conditions were selected by the way to keep temperature and reducing power appropriate for LNT operations. That target resulted in a quite long lean period with 5-15 s rich periods ($\lambda \sim 0.95$). The focus by the engine point selection was at low temperatures important in New European Driving Cycle (NEDC). The emissions after engine, DOC+DPF, LNT and SCR were measured by the sample gas collected to the FTIR analyzer (Gasmeter) calibrated for exhaust gas components. λ values and temperatures were detected also in these positions.

Engine-Bench Experiments for Aftertreatment System Development

The full-scale experiments for investigating the NO_x purification behavior of the combination system as well as the performance of the fuel reformer were conducted with a high efficient 1.4 L 3-cylinder Diesel engine on a transient engine test bench. The investigated systems (LNT+SCR, fuel reformer) were placed under-floor downstream the serial close-coupled DOC+DPF unit and low-pressure EGR branch. The catalysts were thermally conditioned by several hours of DPF regeneration operation. The control logic of the EATS, which was completely new developed, was running on a rapid control system independently from the engine. A few necessary informations were transferred between engine and EATS control via ECU bypass. For steady-state investigations, the engine operation (speed, load, operation mode) was set in order to adjust target conditions at the LNT (temperature, NO_x concentration, exhaust mass flow, A/F ratio). Additionally, transient test cycles (NEDC, WLTP, FTP75, CADC, highway, ADAC) were conducted in order to evaluate and calibrate the system performance and operation strategy. The emissions were measured by different analyzers (CLD: NO_x, NO; NDIR: CO, CO₂; Magnos: O₂; FID: THC; FTIR: NO, NO₂, CO, CO₂, H₂O, NH₃, N₂O, CH₄; LDS: in-situ NH₃; mass spectrometer: H₂) and sensors (NO_x, λ) at various positions (engine out, downstream DPF, upstream and downstream SCR). The gas composition of the out-line exhaust gas fuel reformer was calculated based on emission measurements in the full exhaust gas flow and the dilution factor. The fuel reformer functionality could be demonstrated on engine test bench [4], but is not discussed here. Based on the achieved on-board reformate gas composition, a reformate substitute gas was applied in parallel, containing molar fractions of 20 % H₂, 20 % CO and 60 % N₂.

Vehicle Experiments

A Golf Variant BMT with a dedicated 3-cylinder TDI® engine with a capacity of 1.4 L was applied as the base vehicle, where the developed EATS was built in. In order to reach the highest efficiencies and the lowest CO₂ emissions a variety of technical measures have been tested and applied. The emissions (CO₂, NO_x, CO, THC, PM) were evaluated by the standard New European Driving Cycle (NEDC) containing low-speed urban and high-speed Extra Urban Driving Cycles (EUDC). Alternative fuels to diesel were also investigated in this project.

RESULTS AND DISCUSSIONS

Catalyst Development

Laboratory Simulations

The catalyst development for LNT and SCR as well as the effect of lean-rich conditions were analyzed widely in our earlier study [5], where platinum in PtRh LNT catalyst was

shown to dominate the LNT functionality. Platinum as an active metal catalyzes NO oxidation to NO₂ which is adsorbed on NO_x adsorbents like Barium. Rhodium has a minor role to promote rich side reactions during the enrichment peaks. In laboratory simulations, the duration and λ values were selected to correspond to the real transient conditions created by engine or reformer enrichments. It is important that the reduction power was similar in simulations like in engine conditions. Therefore, the common duration of rich peak in simulations was about five seconds, which was possible to repeat exactly in laboratory equipment. The longer rich periods affect the predicted fuel economy in full-scale, which is also a key part of system optimization. It is not possible to compensate longer rich periods by longer lean periods, because the NO_x adsorption capacity is the limiting factor in lean.

Oxygen storage capacity (OSC) has an active role also in PtRh/LNT catalysis and NO_x conversions were enhanced in the presence of an optimized amount of oxygen storage capacity. It may be assumed that OSC prevents ammonia formation on LNT due to buffering adsorbed oxygen but the results showed opposite trends. Ammonia formation was higher on the LNT concepts having the optimized amount of OSC compared to the OSC-free coatings. This is related to the overall reduction efficiency of NO_x, where ammonia was formed in a side reaction with the low λ values below 1.

The durability of developed catalysts was investigated and confirmed by earlier hydrothermal (HT) durability studies [5]. The catalysts in the experiments stand well HT700/HT800 ageing without deactivation. More severe ageing will happen after ageing at 800-900°C. Thus, fresh catalysts were used in reaction studies for the simplicity.

LNT+SCR Simulations

After screening the durable SCR catalysts with a high NH₃ adsorption capacity and optimizing the PtRh/LNT composition [5], these samples were applied in LNT+SCR simulation with synthetic feed gases. NO_x conversion and the emissions of NH₃ and N₂O were detected in the comparison between LNT only and LNT+SCR. LNT only resulted in high mean NH₃ formation in the simulation with $\lambda=0.82$ in rich (5s). These high NH₃ emissions left a lot of potential to utilize SCR after LNT, which was good for reaction studies. Both copper and iron based SCR catalysts were able to cut NH₃ down and increased particularly the low temperature activity, which is important for passenger car applications. The total volume was fixed in the comparison to LNT only (total 30.000 h⁻¹). When the SCR unit was added after the LNT only, the NO_x conversion were higher and NH₃ slip still lower but that design meant 50% larger total volume [5]. In this simulation, N₂O formation was related solely to HC-SCR (propene as HC) in lean side, no side reaction to N₂O was detected during enrichment peaks.