

Radio-Frequency-Based Urea Dosing Control for Diesel Engines with Ammonia SCR Catalysts

Markus Dietrich, Carsten Steiner, Gunter Hagen, and Ralf Moos
 University of Bayreuth

ABSTRACT

The radio-frequency (RF) or microwave-based catalyst state determination offers the opportunity to operate an automotive catalyst at its optimal point. This has already been proven for the oxidation state of TWCs, the soot loading state on DPFs/GPFs, and the ammonia storage state of vanadium and zeolite based SCR catalysts. However, the latter has only been demonstrated in laboratory scale with synthetic exhaust using gaseous ammonia.

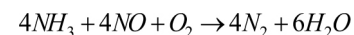
This work presents first results on an engine test bench with a serial-type zeolite-based SCR catalyst, using urea solution and the RF tool to detect the current ammonia loading in real time and to control directly the urea dosing system without any additional sensors. The original catalyst volume was reduced by 50 % to operate deliberately the SCR system under high space velocities and to challenge its function. Stationary conditions and operation points with continuously changing NO_x emissions and space velocities were observed. In all tests, high NO_x conversion was achieved and the ammonia loading could be detected reproducibly in all states by the RF tool. Furthermore, the RF tool was successfully used with closed-loop control of the urea dosing as a two-point control with and without hysteresis. By varying the controlled ammonia storage window, the operation borders of too high or too low ammonia storage were investigated, and the ideal storage value was found. The performed experiments demonstrate that direct operation on a specific ammonia loading can ensure both maximum NO_x conversion and avoid ammonia slip, even with space velocities over 180,000 h⁻¹. Therefore, the control strategy using the RF tool might allow a catalyst volume reduction.

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INTRODUCTION

Current developments in worldwide emission legislations in exhaust gas aftertreatment for diesel engines demonstrate that the reduction of nitric oxides (NO_x = NO + NO₂) of diesel engines is one of the most challenging factors for automotive manufacturers. Within the last years, the ammonia (NH₃) based selective catalytic reduction (SCR) technology has become the major strategy to meet the continuously tightening emission limits for low and heavy-duty diesel engines. The upcoming replacement of the test cycle based emission certification by real-world driving emission (RDE) tests in Europe is the next milestone [1]. Therefore, new concepts for direct catalyst control are required for aftertreatment system optimization.

In NH₃-SCR systems, an aqueous solution of 32.5 wt% urea in water (DEF = diesel exhaust fluid, AUS 32 = aqueous urea solution, or AdBlue™) is injected into the exhaust. After water evaporation, the urea decomposes and forms gaseous NH₃. The latter adsorbs on the active sites of the SCR catalyst and reduces NO_x to water (H₂O) and nitrogen (N₂) [2, 3]. Depending on the NO to NO₂ ratio of the exhaust, different SCR reactions occur in the catalyst. There are two main reactions: the standard SCR reaction (Eq. 1) and the fast SCR reaction (Eq. 2) with equimolar amounts of NO and NO₂ [2, 3].



(1)



(2)

For high NO_x conversion, a specific NH₃ surface coverage of the catalyst is desired, based on catalyst type and its reaction kinetics. Vanadia-based SCR catalyst are established, but current developments target zeolite based materials with Iron or Copper as active components. Current engine operation strategies aim to save fuel and reduce CO₂ emissions resulting in lower exhaust temperatures [1]. Due to their higher low temperature activity Copper exchanged zeolites receive today's most attention [3, 4, 5]. Besides its necessity for the SCR reactions, the previous adsorption of NH₃ on the catalyst surface is required to buffer the continuous changes of operation conditions [2]. This leads to the conflict of high enough NH₃ surface coverage to secure high NO_x conversion and to avoid any NH₃ slip downstream of the catalyst [6]. The latter is also limited in the newest emission legislations [7].

In current SCR systems, the fully model-based urea dosing control approximates the current NH_3 storage relying on gas sensors (NO_x and/or NH_3 sensors) and the urea dosing module [8, 9, 10, 11, 12]. Model uncertainties and the possibility that too high NH_3 storage and an instantaneous catalyst heating may cause rapid NH_3 desorption are leading to the disadvantage that most of today's SCR catalysts are oversized. The use of ammonia slip catalysts (ASC) to oxidize possible downstream NH_3 is also common. However, as a consequence, additional NO_x by the NH_3 oxidation might be formed [3, 9]. The function of the urea-dosing module and the urea concentration need to be diagnosed, since the SCR reaction and the NH_3 storage models rely on their data [13, 14]. A direct measure of the NH_3 storage for urea dosing control avoiding all these possible error sources is still not available.

Within the last years, several studies to determine the state of automotive catalysts *in-situ* using a contactless radio-frequency-based approach were performed [15, 16]. This suggested technique uses the metal canning of the catalyst or filter to form an electrical cavity resonator. Dielectric changes of the catalyst or filter material related to its state can be extracted by analyzing the resonance parameters of the cavity. Successfully proven applications are the determination of the oxygen storage of the three-way (TWC) catalytic converters [16, 17, 18, 19], or the soot loading of diesel particulate filters (DPF) [20, 21]. A combined system of a TWC coated gasoline particulate filter (GPF) has also successfully been monitored under transient conditions within the European driving cycle (NEDC) [22]. The ability of this technique to measure the storage of NO in lean NO_x traps (LNT) [23] is proven, but suffers from a small signal. In SCR applications, the proven ability to monitor the stored amount of NH_3 on the catalyst surface for Vanadia- [24] and especially zeolite-based SCR catalysts [25, 26, 27] receives more and more attention.

RADIO-FREQUENCY CATALYST MONITORING

The basic principle of the RF approach is the so-called cavity perturbation method. This technique is based on the excitation of standing electromagnetic waves (resonances) inside a hollow metal cavity, which in this case is the catalyst canning. Besides the basic geometry of the cavity, its filling (catalyst or filter) interacts with the electromagnetic field and affects the resonance properties. Each resonance can be fully described by two resonance parameters: the resonance frequency f_{res} and the unloaded quality factor Q_0 . These two parameters are related to two different properties of the catalyst material. The change of the resonance frequency $\Delta f_{\text{res}}/f_0$ is directly connected to polarization effects mirrored in the change of the real part of the dielectric permittivity $\Delta \epsilon_1$ (Eq. 3). Effects related to conductivity and dielectric losses are quantified in the change of the imaginary part of the dielectric permittivity $\Delta \epsilon_2$ and are measurable in the change of the reciprocal unloaded quality factor ΔQ_0^{-1} (Eq. 3). A

more detailed description of the measurement technique, the used assumptions and the theoretical background for the extraction of the resonance parameters can be found in previous work [22, 28, 29, 30].

$$\frac{\Delta f_{\text{res}}}{f_0} \propto \Delta \epsilon_1 \quad (3)$$

$$\Delta Q_0^{-1} \propto \Delta \epsilon_2 \quad (4)$$

Similar studies focusing merely on the material effects of powder samples of zeolite SCR catalyst materials already have proven that both ϵ_1 and ϵ_2 are suitable to monitor the NH_3 loading on SCR catalysts with a linear correlation. It is noteworthy to mention that the sensitivity of the material to NH_3 and its cross-sensitivities, especially to humidity are dependent to the catalyst material and its structure. Nevertheless, all observed zeolite-based SCR active materials showed a similar response to NH_3 [29, 31]. The expected material effects of the zeolite to NH_3 is the polar nature of the NH_3 molecule itself and an increase in proton mobility inside of the zeolite structure. Their effects to the conduction mechanisms are also under investigation by impedance spectroscopy (IS) combined with diffuse reflection infrared Fourier transform spectroscopy (DRIFTS) to correlate the electric effects with the reaction mechanism and possible intermediate formation [4, 30, 32].

The excitation of resonances and the extraction of their resonance parameters require coupling elements, which are in this case coaxial probe antennas. By using only one antenna, the one port measurement is limited to the analysis of one reflection parameter. With a second antenna, the number of analyzable signals increases to two reflection and two transmission parameters. The here-shown RF analysis bases on the use of two antennas and on the transmission signal.

EXPERIMENTAL

In this work, a serial-type iron zeolite SCR catalyst on cordierite substrate of PSA was investigated to prove the ability of the RF approach for NH_3 storage determination on the engine test bench and to show its possible benefits in SCR catalyst control. The sample has been extracted of a new serial type exhaust pipe. The used engine test setup is illustrated in Figure 1 and is described in the following. The dynamometer test bench is equipped with a turbocharged 4-cylinder and 2.1 ltr diesel engine (Daimler OM 651, 150 kW), followed by the serial device diesel oxidation catalyst (DOC) and DPF. Then the DEF dosing module (Bosch Denoxtronic 3.2), an uncoated cordierite substrate to support NH_3 formation [33], and a plate mixer are located. One of the two extracted catalyst monoliths with a length of 10.5 cm (volume of 1.7 ltr) was placed in the middle of the RF canning ($\varnothing 5.66''$, length 40 cm) with one RF antenna up- and one

downstream of the catalyst. The geometry of the canning was defined to a perfect cylindrical shape by two inserted metal screens (coarse metal grid) up- and downstream. To determine the catalyst temperature, thermocouples were placed up- and downstream of the canning. Two NO_x sensors were used as an inexpensive alternative for an exhaust NO_x and NH_3 analyzer to determine the current catalyst state and refer it to the RF signals: one sensor was located between the DPF and the DEF dosing, the second NO_x sensor was placed downstream of the SCR canning. For dosing control, no NO_x sensor was used. Additionally, two 50 Ω coaxial cables and the network analyzer (MS46322A, Anritsu) were set up for the RF measurements (not shown in Figure 1). The catalyst has been degreened in a previous experimental run for seven hours at temperatures between 200 and 600 $^{\circ}\text{C}$ before the following experiments were conducted.

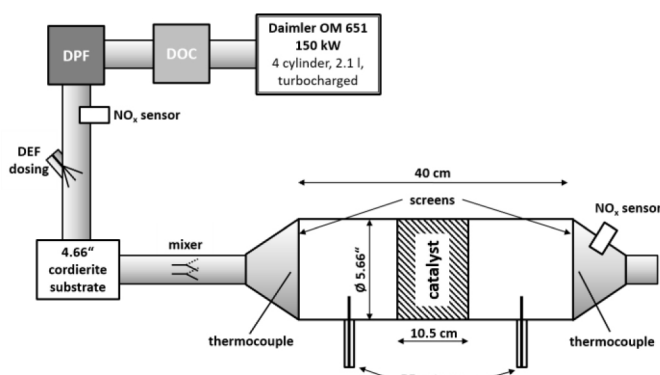


Figure 1. Illustration of the engine setup, in order of gas flow: engine, diesel oxidation catalyst, diesel particulate filter, upstream NO_x sensor, DEF dosing module, uncoated 4.66" cordierite substrate, plate mixer, SCR catalyst canning with thermocouples up- and downstream, two coaxial probe antennas, metal screens, and downstream NO_x sensor.

In the presented study, the engine was operated on four different stationary conditions (engine speed and load), always with a SCR catalyst temperature between 280 and 300 $^{\circ}\text{C}$. The reduced catalyst volume lead to higher space velocities (SV) between 85,000 and 185,000 h^{-1} . The engine out NO_x emissions were varied between 160 and 1,000 ppm in stationary operation points. One additional operation point with continuously changing exhaust gas recirculation (EGR) rate was chosen, resulting in a more dynamic condition with strong changes in space velocity and NO_x emissions.

Example spectra of the analyzed TE_{111} resonance mode (lowest appearing resonance at around 1 GHz) are illustrated in Figure 2 at stationary engine conditions at 300 $^{\circ}\text{C}$ catalyst temperature for an empty (black) and an NH_3 loaded catalyst state (red dashed). One can clearly see the shift of the resonance frequency to lower frequencies, the decrease in peak height and the broadening of the resonance peak resulting from NH_3 storage, as expected from previous work [25, 26, 27,29]. The applied software for real time RF analysis uses complex transmission data and a complex fitting approach for precise f_{res} and Q_0 determination, fully described in [22, 34, 35].

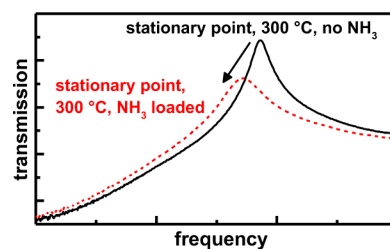


Figure 2. Transmission spectrum of the observed TE_{111} mode at stationary engine conditions at 300 $^{\circ}\text{C}$: without NH_3 (black) and loaded with NH_3 (red dashed).

The control flow diagram of the closed-loop RF urea dosing control used in this study is displayed in Figure 3. Starting at the urea dosing, which requires a definition of the desired DEF dosing rate, based on dosing valve opening times and the pressure of the DEF supply, the thereby formed NH_3 interacts with the SCR catalyst and changes the dielectric behavior of the resonator. The network analyzer measures the complex transmission inside the catalyst canning with an acquisition time of 1.4 s per frequency sweep, which is analyzed and interpreted in real time into the two RF parameters f_{res} and Q_0 . A simple comparator with two-point-control (with or without hysteresis), compares one chosen RF parameter with defined control borders, and starts or stops the urea dosing automatically. In the presented experiments, Q_0 is used for automatically dosing, whereas experiments with f_{res} were also performed. Besides the closed-loop control, a manual dosing control is also possible.

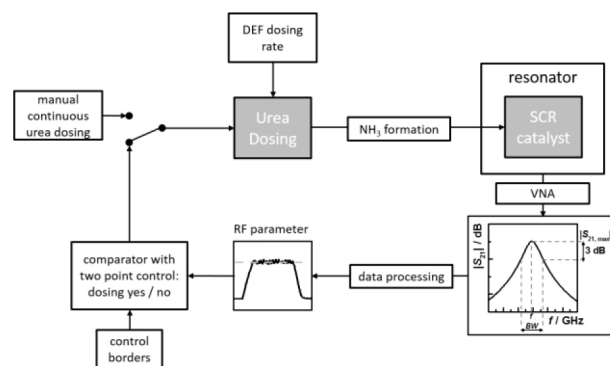


Figure 3. Control flow diagram of the applied RF controlled urea-dosing system.

RESULTS AND DISCUSSION

Since this was the first study performed with the described setup, its functionality and the influence of the DEF dosing rate needed to be investigated. Therefore, the following experiment with a space velocity of 130,000 h^{-1} , $\lambda = 1.35$ and a catalyst temperature of 300 $^{\circ}\text{C}$ was performed, displayed in Figure 4. It shows a) the signals of the upstream (black) and the downstream (red) NO_x sensor; b) the calculated upstream NH_3 concentration based on the DEF dosing rate and the exhaust mass flow; c) the calculated amount of stored NH_3 on the catalyst in gram per liter catalyst volume; d) the change of the resonance frequency $\Delta f_{\text{res}}/f_0$ in reverse scale and e) the change of the reciprocal unloaded quality factor ΔQ_0^{-1} .