

Microwave-Based Catalyst State Diagnosis - State of the Art and Future Perspectives

Ralf Moos
Univ. of Bayreuth

ABSTRACT

The state of catalysts and filters plays a key role in automotive exhaust gas aftertreatment. The soot or ash loading of particulate filters, the oxygen loading degree of three-way catalysts, the amount of stored ammonia in SCR catalysts, or the NO_x loading degree in NO_x storage catalysts are important parameters. Today, they are determined indirectly and/or model-based, calibrated by gas sensors installed up- or downstream of the catalysts or by a differential pressure sensor.

This contribution overviews a novel approach to determine directly the catalyst state by a microwave-based technique. For that purpose, the catalyst housing serves as a cavity resonator. As "sensing" element, one or two simple antennas are mounted in the catalyst canning. The electrical properties of the catalyst device, i.e., of the ceramic honeycomb incl. coating and storage material, can be measured. Such contactless-obtained information is very well correlated with the catalyst or filter state as is demonstrated for different exhaust gas aftertreatment systems.

A comparison that calculates the relative sensitivity concludes this review.

CITATION: Moos, R., "Microwave-Based Catalyst State Diagnosis - State of the Art and Future Perspectives," *SAE Int. J. Engines* 8(3):2015, doi:10.4271/2015-01-1042.

INTRODUCTION AND TECHNICAL BACKGROUND

The legislative pressure on automotive manufacturers to reduce CO₂ emissions lead to booming market shares for diesel engines and leanly operated gasoline engines. Since they are operated leanly, nitrogen oxides (NO_x) cannot be removed with conventional three-way catalysts (TWCs), e.g. [1]. Novel exhaust gas aftertreatment concepts have emerged for that purpose.

For heavy duty vehicles and passenger cars, selective ammonia catalytic reduction systems (NH₃-SCR) have been serialized in the past few years [2,3]. In NH₃-SCR systems, a urea water solution is injected into the exhaust. By hydrolysis, it forms ammonia (NH₃) which serves as a selective reduction agent for NO_x. According to the reaction mechanisms, NH₃ is initially adsorbed (stored) in the SCR catalyst. NO_x conversion depends strongly on the amount of stored NH₃, especially at low temperatures [3,4].

NO_x storage catalysts (NSC), also known as lean NO_x traps (LNT), adsorb NO_x, in a lean phase. NO_x is stored in the form of nitrates. In short rich phases, which follow the longer lean phases, nitrate reduction occurs [5]. To avoid NO_x slip, a direct measure of the amount of stored NO_x would be beneficial. For both NO_x abatement techniques, NH₃-SCR and NSC, a direct measure of the actual catalyst state (stored amount of NH₃ or NO_x, respectively) would be helpful.

Diesel particulate filters (DPF) reduce both the emitted particulate number and mass. To avoid clogging, they have to be regenerated regularly or continuously to burn-off sorbed soot [6]. Regeneration processes consume fuel, and the number of regenerations has to be kept as low as possible. To avoid overloading, it would be beneficial to know precisely the actual soot loading.

Exhausts of conventional gasoline-operated internal combustion engines are treated by TWCs. Only with an average air-fuel ratio (λ) of approximately 1, all limited exhaust components can be converted to almost 100%. λ -probes are installed upstream and downstream of the TWCs to determine the actual oxygen partial pressure of the gas phase [7]. The TWC washcoats contain large quantities of doped ceria-zirconia solid solutions. Since the two different oxidation states of ceria allow storing oxygen, rich/lean fluctuations are buffered. The oxygen storage capacity (OSC) of TWCs is directly related to the quantity of available ceria [8]. Today, the oxidation degree of ceria (oxygen loading state) is calculated indirectly and model-based using data of the λ -probes up- and downstream of the TWCs.

Up to now, classical more or less selective exhaust gas sensors like λ -probes (EGO), wideband oxygen sensors (UEGO), NO_x sensors, or recently NH₃ sensors, are applied to determine the state of the catalysts [9, 10]. In case of DPFs, differential pressure sensors are used, supported by models or to validate models. All these mean

determine the catalyst state only indirectly. “State” in this respect means, e.g., the oxygen loading of TWCs, the NO_x loading of NSCs, the NH_3 loading of NH_3 -SCR catalysts, or the soot loading of DPFs.

A research direction that emerged recently overcomes this indirect gauging. The catalyst state is determined directly by monitoring the electrical properties of the catalysts' coatings themselves. It can be classified in two sub-categories: in a non-contactless approach, and in a contactless, microwave-based one. In the first approach, impedance or resistance measurements of sections of the catalyst coatings are measured. Despite promising results are obtained for TWC [11], LNT [12], and DPF [13], these principles require additional sensor devices and provide therefore no real benefit compared to gas sensors.

MICROWAVE-BASED MONITORING OF EXHAUST GAS AFTERTREATMENT SYSTEMS

A by far higher potential for application has the contactless microwave-based approach. The microwave-based catalyst state diagnosis technique bases on the direct dependence of the electrical properties of the catalyst materials on their loading states [14]. The metallic catalyst canning serves as an electrical cavity resonator. As “sensing” element, one or two simple antennas (e.g., coaxial feed stubs) are mounted in the catalyst housing. The electrical properties of the catalyst device (ceramic honeycomb incl. coating and storage material) can be measured. For catalysts, most work has been done using the resonance behavior, e.g., the resonance frequency obtained from the reflection coefficients (see below) as the signal feature. Alternatively, the power transmission can be used as a signal, which however requires two antennas. The latter principle is used for DPF soot (and ash) loading determination. It will be demonstrated in the present overview that such contactless-obtained information correlates very well with the catalyst state for all above-introduced catalyst types. This will be shown for the determination of the oxygen loading of TWCs, for the amount of stored NH_3 in SCR catalysts, for soot loading of DPFs, and also briefly for the NO_x -loading of NSCs.

Physical basis for the effects in the gas-storing catalysts are changes in the dielectric losses and/or in the permittivities of the catalysts' coatings when being loaded with NO_x (NSC), NH_3 (SCR), or oxygen (TWC). In the case of DPF, the conductive soot causes strong losses that are mirrored in both the frequencies of the resonance modes and the transmission factors.

The Principle of the Microwave-Based Technique

The basic setup of the microwave-based technique is shown in Fig. 1. One or two coaxial waveguide feeds (“antennas”) are mounted inside the canning. The cavity resonator can be seen as a so-called two-port for microwaves. If electromagnetic waves of the complex amplitudes a_1 are excited by one antenna, and if the reflected complex amplitudes b_1 or the transmitted amplitude b_2 are recorded, the input reflection coefficient $S_{11} = b_1/a_1$ for matched output ($a_2 = 0$) or the transmission coefficient $S_{21} = b_2/a_1$ can be calculated. The latter describes the transmission of energy from one port to the other through the interior of the cavity resonator. More details of the measurement technique itself can be found in literature [15, 16].

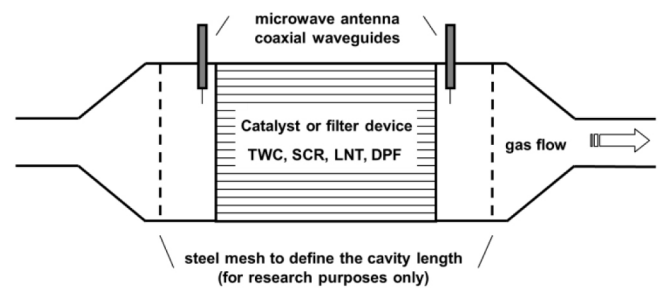


Figure 1. Setup of the microwave-based automotive catalyst state diagnosis. The steel meshes are inserted for research purposes only. They define the cavity length more exactly.

Application for TWC

The basic results of the microwave-based technique are similar for all exhaust gas aftertreatment systems. A resonance spectrum of a TWC in a reduced (rich) and oxidized (lean) state is shown in Fig. 2 (adapted from [17]). The resonance frequency f_{res} , the magnitude of the reflection coefficient $|S_{11}|$ at the resonance frequency, or the full width at half maximum (FWHM) are appropriate signal features that correlate with the oxygen loading of the TWC. In the following, only the resonance frequency shifts are considered for the catalyst devices.

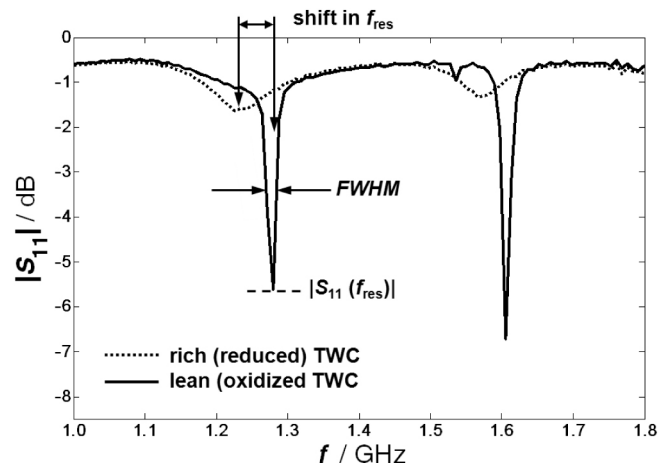


Figure 2. Spectra of the reflection coefficient $|S_{11}|$ of a TWC in the reduced and in the oxidized state in the frequency range from 1.0 to 1.8 GHz. $T = 450^\circ\text{C}$, GHSV 60 000 h^{-1} , lean: $\lambda = 1.05$, rich $\lambda = 0.9$. Data adapted from [17]. Further experimental details are given in [17,18].

The power of this method is shown for instance in a titration experiment conducted in a dynamometer test bench (Fig. 3). The experiment started with a rich phase ($\lambda \approx 0.95$) to complete reduction. Then, the engine switched to $\lambda \approx 1$. Every 20 s, short lean pulses (1 s, $\lambda \approx 1.02$) oxidized the TWC stepwise and gradually. The downstream λ -probe signal indicates stoichiometric operation until about $t = 720$ s, when a sharp increase of λ indicates a fully oxygen loaded TWC.

The microwave signal, however, fully reflects the stepwise oxygen loading. The system is neither cross sensitive to other exhaust components, like H_2 , CO_2 , H_2O , or CO nor to gas flow changes [19], but temperature compensation is appropriate, since the resonance frequency is temperature dependent. One reason for the temperature dependency is the geometrical resonator increase with temperature.