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Detection of the ammonia storage of vanadia-based SCR-catalysts by a radio-frequency method

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Summary:
Stricter regulations for nitrogen oxide emissions require an aftertreatment of exhaust gases from biomass combustion plants, such as biogas cogeneration plants. For this reason, SCR systems are used in these applications and their functionality should permanently be monitored during operation to ensure sufficient efficiency at lowest operating costs. The radio-frequency-based state diagnosis of catalytic converters is suitable for this purpose and has already been extensively tested for three-way catalytic converters (TWC) or NO_x storage catalysts (LNT) in the automotive sector.

Keywords: Selective catalytic reduction (SCR), ammonia loading, vanadium-based catalyst (VWT), cavity resonator, radio-frequency

Motivation

After the emissions of nitrogen oxides (NO_x) from vehicles have been increasingly regulated by law in recent years, the exhaust gases from biomass combustion plants, such as pellet heating systems, are now also affected. Therefore, the use of efficient systems for the selective catalytic reduction (SCR) of NO_x becomes more relevant in the future [1].

In the SCR process, ammonia (NH_3), bound in an aqueous urea or NH_3 solution, serves as a reducing agent for the nitrogen oxides, which are converted to nitrogen (N_2) and water (H_2O) on a catalyst. Before the SCR reactions can take place, a previous adsorption of ammonia at the catalyst is essential. For this reason, the catalyst has the property to store a certain amount of ammonia [2].

Therefore, the control of the exhaust gas aftertreatment in many automotive applications is based on the loading status of the catalyst, which is calculated by means of different sensors up- and downstream the catalyst and on the basis of various models [3].

Since biomass combustion systems are usually individual setups, the effort to determine suitable models would be too complex. Accordingly, the direct NH_3 load monitoring of the catalyst by means of radio-frequency technology is a less extensive and complicated alternative compared to the usual techniques.

Methods and Setup

In the radio-frequency-based state diagnosis, the catalyst itself operates as a sensor. For this purpose, the catalyst housing is regarded as a cavity resonator. The catalyst inside of the metallic housing serves as a dielectric material. It changes its complex permittivity due to NH_3 loading [4]. In addition, the housing contains two openings for the installation of two coupling elements (antennas). The schematic structure is shown in Fig. 1.

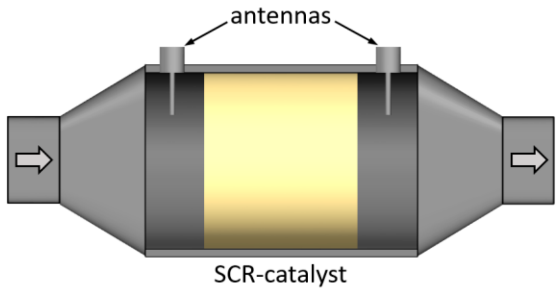

Fig. 1. Schematic design of the catalyst housing as a cavity resonator with built-in antennas

A vector network analyzer is used to couple electromagnetic waves into the catalyst housing via the antennas. Standing electromagnetic waves can be excited at specific frequencies.

The scattering parameters S_{ij}, which represent the reflection and transmission behavior of the catalyst, are used as the measured variable. Depending on the NH_3 loading of the catalyst, its radio-frequency properties change, which results in a shift in the resonance frequency f_{res} and a change in the quality factor Q [5].

Results and Discussion

The following measurements were carried out with a vanadia-tungsten-titanium-based catalyst (VWT; catalyst with 1.7 wt% V_2O_5) at a mixing unit for synthetic exhaust gases (5 % O_2, 5 % H_2O in N_2), while the catalyst was heated to an operating temperature of approx. 400 °C. Meanwhile, the SCR catalyst was loaded with 300 ppm ammonia in the exhaust gas at constant flow rate and unloaded as soon as the maximum was reached. The following investigations refer to the transmission factor S_{21}.

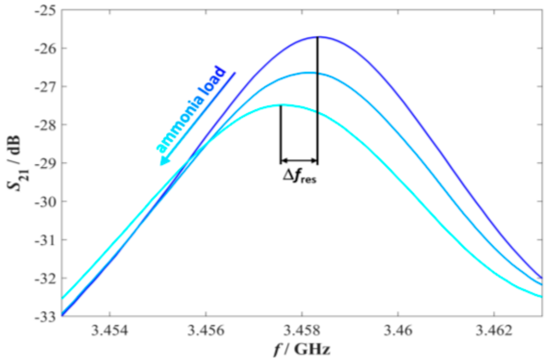

Fig. 2. Scattering parameter S_{21} as a function of frequency for different ammonia loadings of the catalyst at 400 °C

Fig. 2 shows the curves of three transmission spectra at different loading states of the catalyst. The loading of the catalyst with ammonia leads to changes in the electrical properties, including the permeability ε_r and the conductivity σ, which in turn influence the radio-frequency behavior [6]. The measured signal shows a decrease of the resonance frequency f_{res} with increasing ammonia loading.

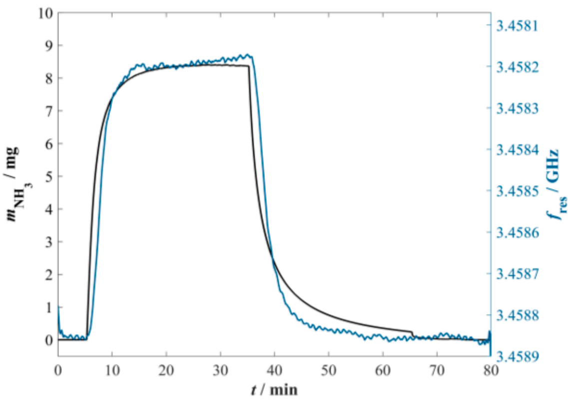

Fig. 3. Comparison of the calculated mass loading m_{NH3} of the catalyst with the measured resonance frequency f_{res} at 400 °C

Using the ammonia concentration up- and downstream of the catalyst determined by an FTIR spectrometer, the amount of stored ammonia can be calculated from the difference and compared with the resonance frequency determined from Fig. 2.

The results (see Fig. 3) show that the two measured values correlate. During loading at the beginning of the measurement, a clear drop in the resonance frequency can be observed (inverted axis) and reaches an almost constant value as soon as the catalyst is fully loaded. After unloading, the resonant frequency returns to its initial value.

Summary and Outlook

It is possible to determine the ammonia loading of catalysts directly by evaluating the resonance frequency of a cavity resonator with a built-in SCR catalyst. The functionality of this simple system was tested primarily during measurements at a gas mixing unit.

Future measurements will be carried out on a real SCR system for the exhaust gas after-treatment of a biogas cogeneration plant. Here, it is important to investigate further information on the cross-sensitivities of this system.

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