Adaption of a capillary sampling technique for spatially resolved concentration measurements in a SCR catalyst under real exhaust gas conditions

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Introduction

The reduction of nitrogen oxides (NO_x) has become increasingly important worldwide over the last decades, particularly for the after treatment of lean-burn engine exhaust gases. In particular in the context of future hydrogen engines, where NO_x are the only emissions of concern, the selective catalytic reduction (SCR) of NO_x is expected to maintain its relevance. By SCR NO_x is reduced to N₂ and H₂O by NH₃ over state-of-the-art Cu-zeolites or vanadia catalysts (VWT). For safety reasons, the reducing agent NH₃ is not supplied as gas but in form of a urea water solution (UWS). The water evaporates and the urea reacts to HNCO and NH₃ by thermolysis. The produced HNCO is then hydrolyzed to NH₃ and CO₂. Due to inhomogenous spray distribution and urea side reactions undesired solid deposits such as biuret, triuret and cyanuric acid can be formed which lead to a decrease of SCR efficiency. In order to enable detailed kinetic investigations on the chemical interactions of UWS with the SCR reaction system, spatially resolved temperature and concentration measurements are carried out in the monolith channels. [1–3]

The invasive capillary sampling technique has been used in various studies and models were developed to predict the hydrodynamics modified by the capillary within the channel [4]. In this work, the sensitive measurement setup is adapted for the use in a hot gas test rig under real conditions. The additional influence of the analytical setup and measurement procedure on the resulting spatially resolved concentration data is quantified in detail.

Adaption of the capillary sampling technique to a hot gas test rig

A setup for capillary sampling was adapted to an existing hot gas test rig including an SCR monolith. Sampling is carried out through a thin capillary, which can be moved through a monolith channel by a motorized stage. FTIR and H₂-MS analytics are

installed downstream the capillary to determine the gas phase concentrations of NH_3 , NO, NO₂, H₂, HNCO, H₂O and CO₂.

Since the sampling flow rate extracted from the monolith channel by the capillary is by two orders of magnitude smaller than the required sample flow rate for the FTIR, a coil setup is designed for sample accumulation. The effect of sample residence time and dispersion within the coil is analyzed. Additional measurements were performed without the capillary and the coil setup in order to quantify the influence of the sampling technique.

Spatially resolved concentration measurements

Reaction kinetic studies were carried out after detailed calibration of the measurement technique in order to improve the understanding of the interactions between urea decomposition and SCR reactions. Both measurements with gaseous NH₃ dosing and with liquid UWS dosing were carried out. In addition to temperature and concentration, the formation of deposits was investigated. Furthermore, the influence of hydrogen on the multiphase reaction system was examined.

Conclusion

A method for quantification of the influence of the sampling technique and analytical setup for spatially resolved concentration measurements under real exhaust gas conditions was developed. Based on this, spatially resolved measurements were carried out to investigate the reaction kinetics of the NH₃-SCR in combination with urea decomposition and subsequent HNCO hydrolysis. Finally, the influence of hydrogen on the reactive multiphase system was evaluated, which is particularly interesting with regard to future hydrogen engines.

References

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