

IN-SITU INVESTIGATIONS OF CATALYTIC NO REDUCTION VIA PLANAR LASER-INDUCED FLUORESCENCE

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Abstract

Visualizing chemical reactions and fluid dynamics inside catalytic reactors non-invasively is feasible using spatially resolved in-situ analytics. The method of planar laser-induced fluorescence (PLIF) can generate two-dimensional species concentration profiles of specific target molecules such as NO and OH. Therefore, the interaction of mass transfer and catalytic surface reaction is directly measurable. In this study, we present two-dimensional species concentration profiles during the catalytic reduction of NO with hydrogen over a Diesel Oxidation Catalyst (DOC) within a channel flow reactor. Experimental conditions such as flow rate, inlet composition, and temperature are varied to elucidate their influence on the catalytic reaction and mass transfer.

Keywords

PLIF, spatial resolution, NO reduction.

Introduction

Spatially resolved in-situ analytics give the opportunity to see what is actually happening inside chemical reactors and catalytic converters. One-dimensional (1D) species concentrations- and temperature-profiles inside catalytic reactors have become available within the last years using different in-situ methods such as capillary sampling techniques, DRIFTS and Raman spectroscopy. The interaction of mass transport and catalytic surface reaction, however, calls for two-dimensional (2D) spatial resolution of the species profiles. In this study we present a recently set-up configuration to determine two-dimensional species concentration and temperature profiles in channel flows over catalytic walls by laser spectroscopy (Zellner et al 2014). Exemplarily, results are shown for the reduction of NO by hydrogen to ammonia above the catalytic channel wall of a Diesel Oxidation Catalyst (DOC) (Zellner et al. 2015). Planar Laser Induced Fluorescence (PLIF) provides absolute 2D-NO gas-phase concentration profiles at

varying external conditions such as flow rate, inlet composition, and temperature.

Experimental

Experiments have been carried out inside an optically accessible flow reactor with a commercial Pt/Al₂O₃ catalyst (147 g/ft³, 400 cpsi). Excitation of NO was performed via laser radiation at 226.68 nm. For this purpose a tunable dye laser was pumped by a Nd:YAG laser for UV laser light generation. Fluorescence of NO has been detected by an ICCD camera with an appropriate band-pass filter. The measured NO fluorescence images have been corrected for inhomogeneities of the laser light sheet, the photo-cathode sensitivity of the ICCD-camera and pulse to pulse intensity fluctuations of the laser beam. Electronic quenching of the excited NO molecules by all relevant gas phase species has been considered.

Additionally, the product gas flow was continuously analyzed by means of online FTIR and mass-spectrometry.

Results

Figure 1 shows absolute quenching corrected NO-concentrations inside the flow reactor under reactive conditions. The length of the catalytic plate is 10 mm. In front of the catalyst, the NO-concentration is homogeneous, no chemical reaction is noticeable. Above the catalytic surface the reduction of NO to NH_3 has a significant influence on the observed NO-concentrations. Gradients in vertical as well as horizontal directions appear, visualizing the catalytic conversion of NO as well as correlated diffusion processes. In vertical direction gradients reveal diffusion processes which are additionally influenced by convection of the gas flow. The impact of operation temperature and flow rate (residence time) on the concentration gradients is illustrated in Figure 2. Increasing the temperature and decreasing the flow rates results in higher NO conversions and thus stronger gradients. NO concentrations obtained by ex-situ FTIR analysis show good agreement with concentrations determined with PLIF at the outlet of the observed area.

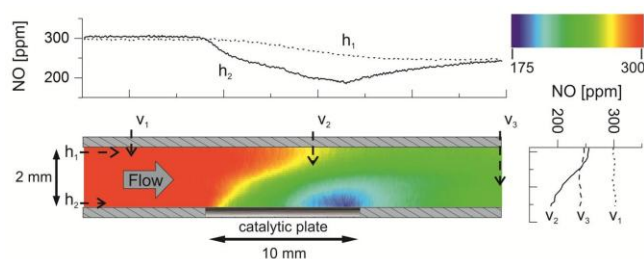


Figure 1. 2D spatially resolved NO-concentration within the flow channel for catalytic reduction of NO to NH_3 over a $\text{Pt}/\text{Al}_2\text{O}_3$ catalyst. $c_{\text{NO}} = 100 \text{ ppm}$, $c_{\text{H}_2} = 1000 \text{ ppm}$, balance N_2 , $T = 300^\circ\text{C}$.

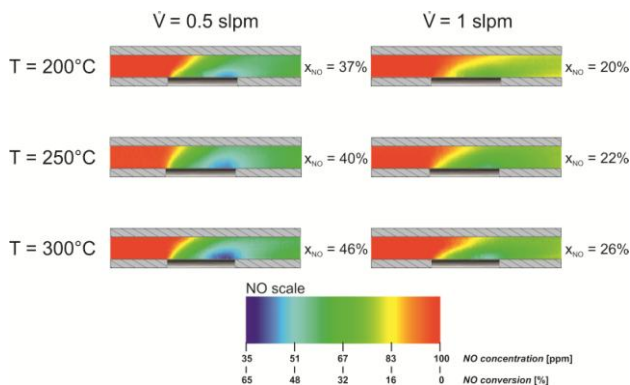


Figure 2. Influence of temperature and volumetric flow rate on the NO distribution. $c_{\text{NO}} = 100 \text{ ppm}$, $c_{\text{H}_2} = 1000 \text{ ppm}$, balance N_2 .

Conclusions

The presented experimental technique provides 2D concentration profiles of gas-phase species over catalytic surfaces to elucidate the interactions between surface kinetics and mass and heat transport.

References

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 Zellner, A., Suntz, R., and Deutschmann, O., *Angewandte Chemie*, 54, p. 2691–2693, (2015)