



Experiment title: Spatially resolved XAS studies for elucidating the role of catalyst composition on the noble metal state during NO_x-removal by SCR with H₂

Experiment number:
CH-6595

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Report:

To reduce the global CO₂ emissions, the use of hydrogen as a carbon-free alternative fuel for lean-burn internal combustion engines is a promising alternative. The only harmful pollutant of such applications is the NO_x formed due to the high combustion temperatures. The removal of NO_x emissions is typically done by selective catalytic reduction with hydrogen (H₂-SCR) over catalyst containing noble metals (primarily Pt or Pd) supported on various mixtures of metal oxides [1]. Depending on the catalyst composition, i.e. support/dopant type, acidic/basic sites or NO_x storage capabilities are generated, which strongly affect the activity profile at low temperatures [2, 3]. Furthermore, challenges such as unselective H₂ reaction with oxygen or the formation of N₂O still need to be solved. Despite numerous catalytic systems have been investigated in previous studies, an in-depth understanding of the reaction mechanism and of the catalyst structure under reaction conditions is missing.

In this context, our study aimed at elucidating the structural peculiarities, synergism and influence of basic (Ba) and acidic (W, Nb) promoters on the H₂-SCR activity of a 0.5 wt.% Pt/CeO₂ catalyst. For this purpose, systematic *operando* experiments were conducted at Pt L₃ edge during catalyst pre-treatment and under different reaction conditions. To reveal structural gradients along the catalyst bed, spatially resolved XAS measurements were performed at three different positions between the reactor inlet and outlet. The XAS data were recorded in fluorescence mode using a quartz capillary *operando* cell in plug-flow geometry that was heated by hot-air gas blower. Reactants (O₂, NO, H₂ in He) were dosed using the gas dosage system available at the beamline. The outlet gaseous products were monitored *via* a mass spectrometer and an FTIR instrument.

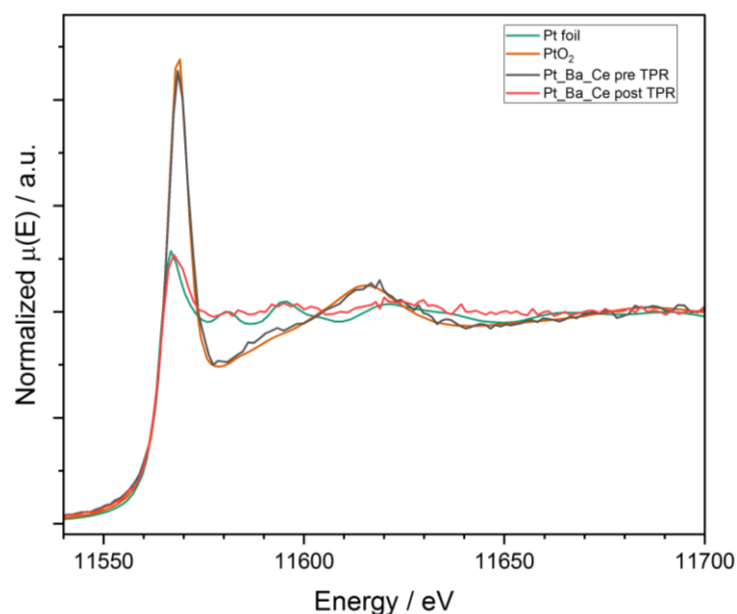


Figure 1: XANES spectra collected at the Pt L₃ edge of a Pt/Ba-CeO₂ catalyst before (as prepared state) and after temperature programmed reduction (TPR) in 5%H₂/He.

Preliminary data analysis indicate the presence of highly dispersed Pt species in the as prepared Pt/CeO₂ catalysts (Fig. 1). During a reductive pre-treatment, metallic Pt is formed, independent of the catalyst promoter. At the middle of the catalyst bed, partial reoxidation was observed during the H₂-SCR reaction for Pt/Ba-CeO₂ catalyst. A more detailed analysis of the spatially resolved *operando* XAS data is performed currently at KIT to further elucidate the correlations between catalyst structure and activity.

References:

1. Y. Guan et al., J. Environ. Chem. Eng., 2021. **9**(6): p. 106770.
2. F. J. P Schott et al., Appl. Catal. B, 2009. **87**(1): p. 18-29.
3. C. N. Costa and A. M. Efstathiou, Appl. Catal. B, 2007. **72**(3): p. 240-252.