



**Experiment title:** Spatially resolved XAS studies for elucidating the role of catalyst composition on the noble metal state during NO<sub>x</sub>-removal by SCR with H<sub>2</sub>

**Experiment number:**  
CH-6595

<b>Beamline:</b> BM23	<b>Date of experiment:</b> from: 02/07/2023 to: 10/07/2023	<b>Date of report:</b>
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## Report:

To reduce the global CO<sub>2</sub> 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<sub>x</sub> formed due to the high combustion temperatures. The removal of NO<sub>x</sub> emissions is typically done by selective catalytic reduction with hydrogen (H<sub>2</sub>-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<sub>x</sub> storage capabilities are generated, which strongly affect the activity profile at low temperatures [2, 3]. Furthermore, challenges such as unselective H<sub>2</sub> reaction with oxygen or the formation of N<sub>2</sub>O 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 H<sub>2</sub>-SCR activity of a 0.5 wt.% Pt/CeO<sub>2</sub> catalyst. For this purpose, systematic *operando* experiments were conducted at Pt L<sub>3</sub> 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 (O<sub>2</sub>, NO, H<sub>2</sub> 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.

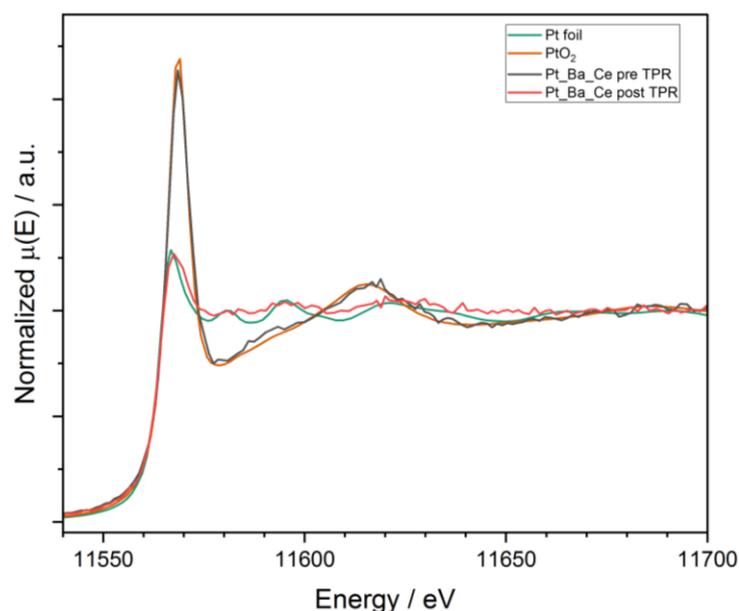


Figure 1: XANES spectra collected at the Pt L<sub>3</sub> edge of a Pt/Ba-CeO<sub>2</sub> catalyst before (as prepared state) and after temperature programmed reduction (TPR) in 5%H<sub>2</sub>/He.

Preliminary data analysis indicate the presence of highly dispersed Pt species in the as prepared Pt/CeO<sub>2</sub> 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 H<sub>2</sub>-SCR reaction for Pt/Ba-CeO<sub>2</sub> 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.