



Experiment title: The role of coordination chemistry and structure of iron in zeolites for selective catalytic reduction of NO_x with ammonia

Experiment number:
CH4406

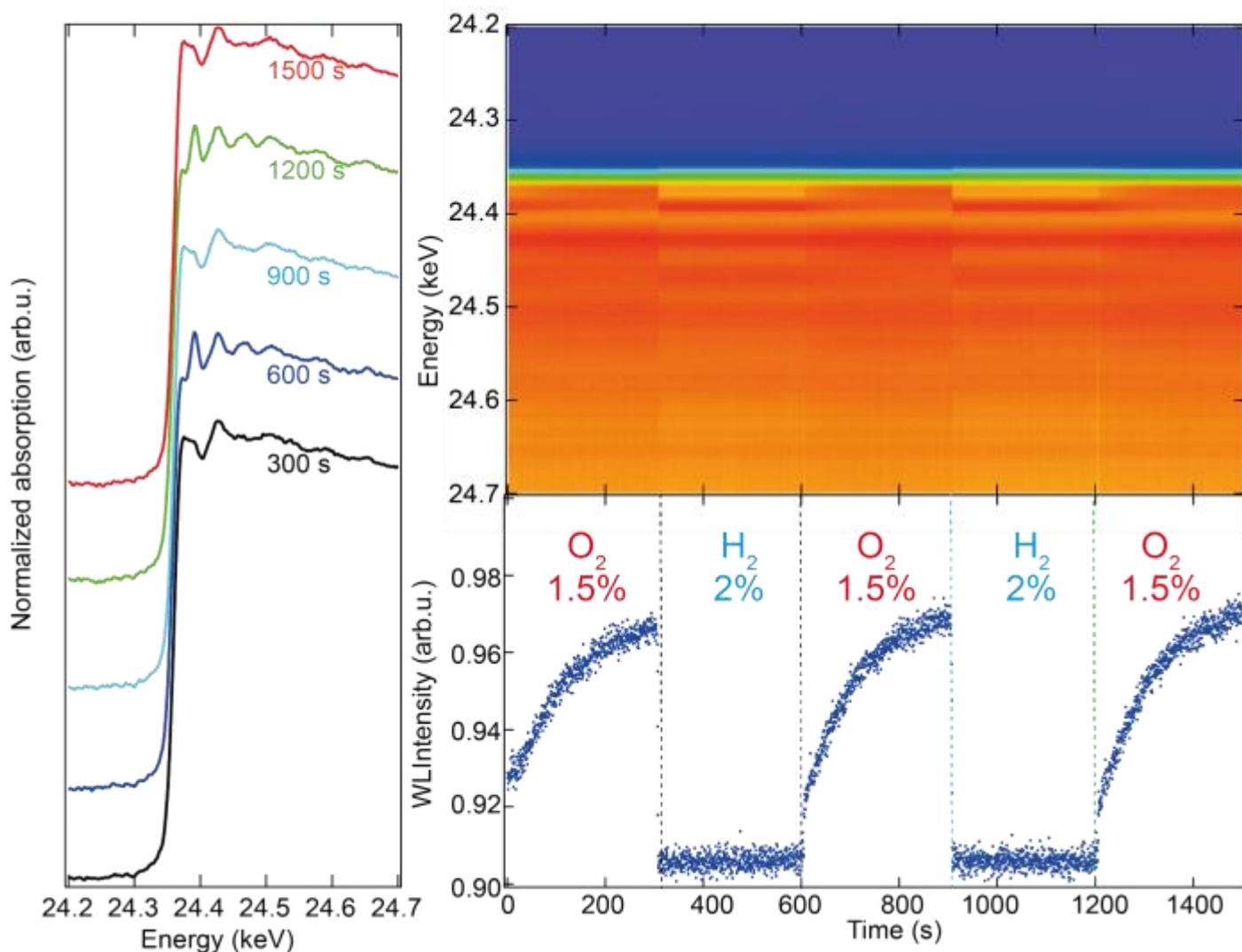
Beamline: ID24	Date of experiment: from: 150624 to: 150630	Date of report: 160914
Shifts: 15	Local contact(s): Olivier Mathon (/Giovanni Agostini)	<i>Received at ESRF:</i>
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Report:

We start this report with a short clarification of the title of the experiment. The CH4406 experiment was accepted for investigating iron functionalised zeolites. It turned out, however, that iron was not possible to study with the present the design of the reaction cell since these elements would require more transparent windows. Instead we were asked to study elements using higher energies. We chosed to study palladium in palladium-based catalysts.

During the experimental session we carried out several *ex situ* and *in situ* ED-XAS measurements with synchronous mass spectrometry (MS). The main focus was on characterising alumina supported Pd and bimetallic Pt-Pd during oxidation-reduction with O₂/H₂ cycling and during methane oxidation by in situ ED-XAS/MS. For the methane oxidation experiments a transient approach was used with a pulsed flow of different oxygen concentrations were performed.

Here we show in Figure 1 the XAS spectra recorded during consecutive 300 s oxidising (1.5 % O₂) and reducing (2 % H₂) periods over Pd-Pt/Al₂O₃ F500 at 360 °C. The XAS spectra (panel a) were recorded at the end of the oxidising and reducing periods. In panel b the color coded intensities of XAS spectra (blue = low intensity, red = high intensity) and in panel c the XAS white line intensity at 24 372 eV is shown. The hysteresis observed in panel c (supported also by other measurements in our home laboratories) suggests that the Pd nanoparticles change not only the oxidation state and local structure but also the entire particle morphology. During the oxidation the particles change shape and expose a larger or more accessible surface so when hydrogen is introduced the reduction to metallic Pd particles are relatively faster.



The results have been analysed and a manuscript has been submitted to Journal of Physical Chemistry C [1]. Another manuscript on the methane oxidation measurements is composed at present.

Figure 1. XAS spectra recorded during consecutive 300 s oxidising (1.5 % O₂) and reducing (2 % H₂) periods over Pd-Pt/Al₂O₃ F500 at 360 °C. (a) XAS spectra recorded at the end of the oxidising and reducing periods. (b) Color coded intensities of XAS spectra (blue = low intensity, red = high intensity) and (c) the XAS white line intensity at 24 372 eV.

References

- [1] Characterization of Surface Structure and Oxidation/Reduction Behaviour of Pd-Pt/Al₂O₃ Model Catalysts, N. Martin, J. Nilsson, M. Skoglundh, E. Adams, X. Wang, P. Velin, G. Smedler, A. Raj, D. Thompsett, H. Brongersma, T. Grehl, G. Agostini, O. Mathon, S. Carlson, K. Norén, F. J. Martínez-Casado, Z. Matej, O. Balmes, P.-A. Carlsson. *Submitted to Journal of Physical Chemistry*.