



	<b>Experiment title:</b> Time-resolved <i>in situ</i> XAS/FTIR/MS study on methane oxidation over alloyed Pt-Pd supported catalysts	<b>Experiment number:</b> CH3089
<b>Beamline:</b> ID24	<b>Date of experiment:</b> from: 100609 to: 100615	<b>Date of report:</b> 100825
<b>Shifts:</b> 15	<b>Local contact(s):</b> Santiago Figueroa and Mark A. Newton	<i>Received at ESRF:</i> 110217
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### Report:

We have carried out time-resolved *in situ* dispersive XAS/IR/MS measurements on i) alumina and ceria supported Pt-Pd catalysts and ii) three alumina supported Pd catalysts with different dispersions (i.e., Pd crystallite size) during methane oxidation at transient conditions. Both temperature programmed experiments (reaction, oxidation and reduction) and reactant step- and pulse response experiments were carried out. The focus was to understand more about the dynamics of the Pd phase as a function of reactions conditions, support material ( $\text{Al}_2\text{O}_3$  and  $\text{CeO}_2$ ), Pd-Pt alloying and Pd dispersion.

During the laboratory session we focused on detailed studies of catalytic activity of methane oxidation. The chemical state of the Pd crystallites was studied in the XANES region for the Pd K absorption edge and correlated with the adsorbate composition following various specific absorption bands (IR spectroscopy) and catalyst activity/selectivity (mass spectrometry). For evaluation of the XANES data we have previously used a method that is based on analysis of the area under the white-line (WLA). This method was developed for analysis of the Pt  $L_{\text{III}}$  edge [1, 2] and thus we are in a process of developing this further for the analysis of Pd K edge, which appears different as compared to Pt  $L_{\text{III}}$  edge.

The included figure shows the results from temperature-programmed reaction experiment where the methane oxidation is studied for both prereduced and preoxidised Pd/ $\text{Al}_2\text{O}_3$  and Pd/ $\text{CeO}_2$  powder samples. The degree of Pd oxidation can clearly be followed via the XANES at the Pd K edge and correlated with adsorbate composition and methane oxidation activity not shown in the figure. At present we are working on two scientific paper on these results [3, 4].

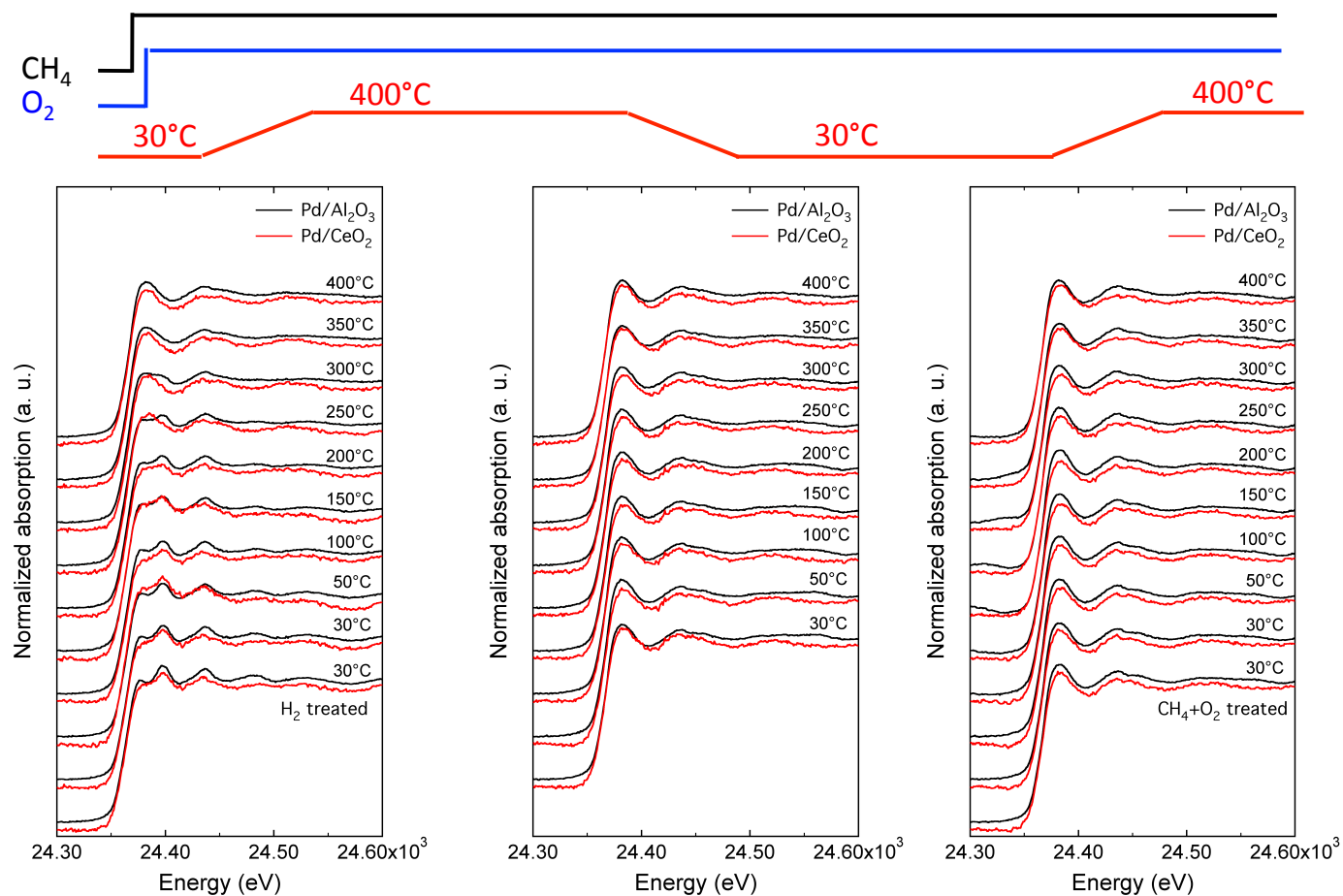


Figure 1. Temperature-programmed reaction of methane oxidation on reduced and oxidised Pd/Al<sub>2</sub>O<sub>3</sub> and Pd/CeO<sub>2</sub> powder samples. The degree of Pd oxidation is followed via the XANES at the Pd K edge and correlated with adsorbate composition (IR spectroscopy) and methane oxidation activity (mass spectrometry) not shown in the figure.

## References

- [1] E. Becker, P.-A. Carlsson, H. Grönbeck and M. Skoglundh, *J. Catal.* **252** (2007) 11.
- [2] E. Becker, P.-A. Carlsson, L. Kylhammar, M. Newton and M. Skoglundh, *J. Phys. Chem. C.* **115**, (2011) 944.
- [3] EDXAS/MS study on CH<sub>4</sub> oxidation over ceria supported Pd and Pt-Pd alloy particles, *to be submitted to J. Catal.*
- [4] EDXAS/MS study on CH<sub>4</sub> oxidation over Pd/Al<sub>2</sub>O<sub>3</sub> catalyst with different Pd dispersion, *to be submitted to J. Phys. Chem. C.*