ESRF	**Experiment title:** Understanding the morphology of aged Pt-Pd alloy catalysts and how that determines the activity for methane oxidation	**Experiment number**: CH4578
Beamline: ID24	**Date of experiment**: from: 160303 to: 160309	**Date of report**: 160319
Shifts: 15	**Local contact(s)**: Debora Motta Meira	*Received at ESRF:*

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Report:

During the experimental session we carried out several *ex situ* and *in situ* ED-XAS measurements with synchronous mass spectrometry (MS).

First a number of *ex situ* measurements on alumina supported Pd-Pt were carried out. These will be used straight away as complementary information in a manuscript on oxidation-reduction behaviour of Pd-Pt, which is based on *in situ* measurements performed at ID24 as well as a number of other methods carried out in our home laboratories during 2015 [1].

Second, we revisited methane oxidation over a series of palladium catalysts with varying Pd dispersion. This was supposed to be finished in the original CH4578 experiment in November 2015 but instead we carried out the full series in March. Time-resolved *in situ* measurements during transient supply of reactants were carried out at several temperatures and several levels of oxygen. The results will be combined with XRD and TEM from our home laboratories in a manuscript during 2016/17.

Third, again we used time-resolved *in situ* ED-XAS/MS but this time to study the impact of water on palladium based catalysts during complete oxidation of methane. This was done by use of a portable water formation reactor (Chalmers equipment) that we brought to the beamline. Due to several issues with the reaction cell (likely as a result of overheating in the first measurement caused by a failing thermocouple) and unstable beam towards the end of the beam time, water impact measurements could just be done on very few samples. Hopefully, we will be able to publish something on this but likely this needs to be complemented in future experiments.

The Figure 1 shows a typical result for an alumina based Pd catalyst with step-response experiment adding/remiving either the oxygen or water. The top panels show the white-line intensity at the Pd k-edge and the bottom pansles show the concentration of the effluent methane and products. Figure 2 shows the XAFS spectra for the diffient gas compositions at steady-state (in the end of each phase)

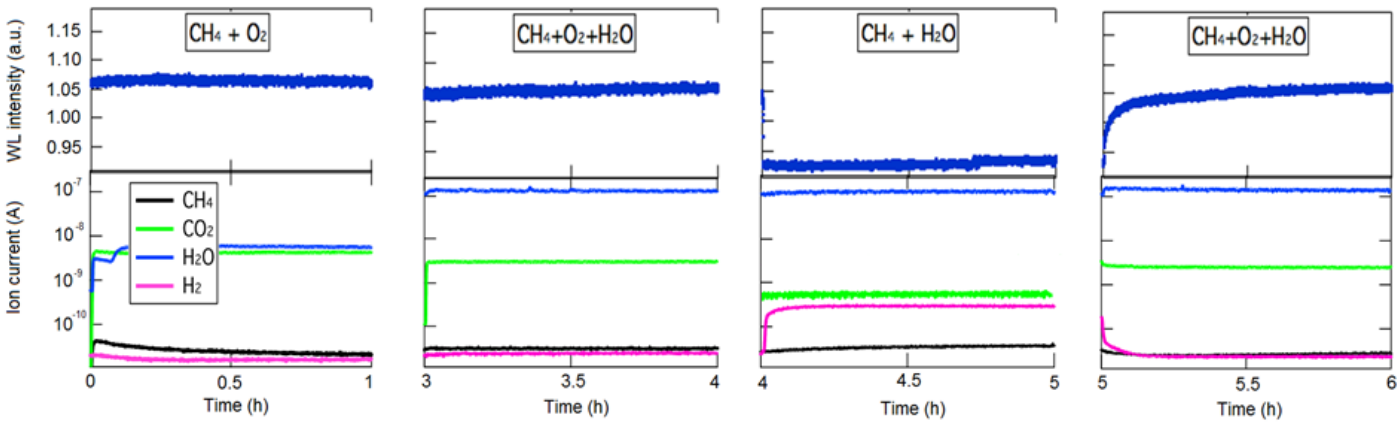

Figure 1. Wite-line intensity at the Pd k-edge (top panles) and concentration of the effluent methane and products (bottom panles) at 380°C during methane oxidation while changing step-wise either the oxygen or water addition.

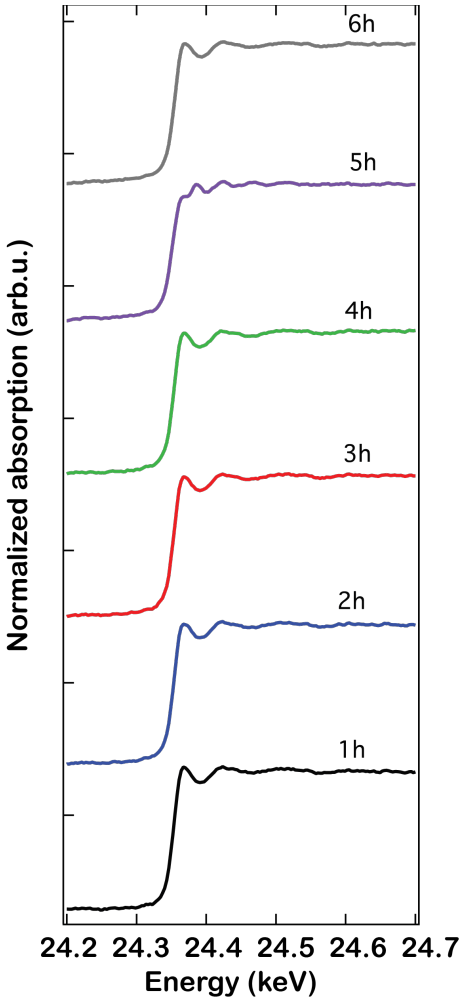

Figure 2. XAFS spectra of the Pd k-edge for the diffient gas compositions at steady-state (in the end of each phase).

References

[1] Characterization of Surface Structure and Oxidation/Reduction Behaviour of Pd-Pt/Al_2O_3 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*