ESRF	**Experiment title:** Time-resolved HXRD/DRIFTS/MS study on transient and SO_2 promoted methane oxidation over alloyed Pt-Pd nanoparticles supported on ceria	**Experiment number**: CH3406
Beamline: ID15B	**Date of experiment**: from: 111116 to: 111121	**Date of report**: 120215
Shifts: 15	**Local contact(s)**: Simon Arthur John Kimber	*Received at ESRF: 120215*

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Report:

During the experimental session we carried out several time-resolved *in situ* HXRD/IR/MS measurements focused on i) methane oxidation over Pt and Pd supported on silica, alumina and ceria as well as Pt-Pd supported on ceria ii) sulfur promoted methane oxidation over alumina and ceria supported Pt and iii) CO oxidation over silica, alumina and ceria supported Pt as well as for pure Co_3O_4. Despite some potentially severe issues with the experimental set-up, which were excellently sorted out by the ESRF scientists M. A. Newton, M. DiMichiel and S. Kimber, we were successful in 15 different types of experiments, each repeated at 3-6 temperatures (in total 57 individual experiments).

Primarily reactant pulse response experiments, i.e., pulsing of O_2, SO_2 etc, were carried out. The focus was to understand more about the dynamics of the active phase (noble metal) as well as the promoter phase (metal oxide support) and the interaction between these under reaction conditions typically relevant for exhaust afterteatment applications.

During the laboratory session we focused on detailed studies of catalytic activity and selectivity of the mentioned reactions. The present experiments will help us to further understand experiments carried out previously at ID24/ESRF. Also some of the results are likely to be combined with results from previous ID24 sessions. For example the transiently high activity we observed during oxygen pulsing over Pt catalysts [1,2] and the behavior of the Pd catalysts (two manuscripts in preparation [3,4] which potentially still may include HXRD results) can be further clarified. Also observed phenomena in other studies on sulfur propomted methane oxidation [5,6], which not involve XAS or XRD, can be better explained with use of the present results.

The included figure shows a typical result from a pulse response experiment obtained during the CH3406 session. Here the methane oxidation is studied over a Pt/ceria catalyst during

oxygen pulsing. The structural changes in both the active and promoter phases can be followed. We will as soon as possible perform deeper analysis of the diffracograms and correlate these with data obtained from the simultaneous infrared spectroscopy and mass spectrometry measurements.

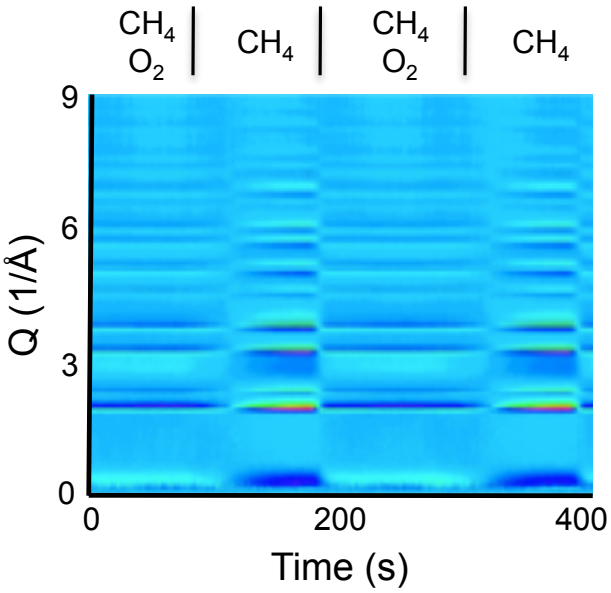

Figure 1. Structural dynamics in the active and promoter phases of a Pt/ceria catalyst during transient methane oxidation at 400°C observed *in situ* with HXRD.

However an unfortunate present bottleneck is to achieve the HXRD data corrected for inherrent oscillations in the detector used at ID15B. To our understanding this can (or should) only be made by the local contact as it is beam line related. We stress this as general input as we think improvement in this area will keep ESRF, which in many aspects is a preferred facility for us, even more competitive/attractive. Also we find this to limit our possibilities to report back to the ESRF and weaken our chances for new proposals.

References

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