ESRF	**Experiment title:** Fighting oxygen poisoning in methane oxidation catalysis	**Experiment number**: CH5632
Beamline: ID24/ID15	**Date of experiment**: from: 181017/ to: 181023	**Date of report**: 230321 *Received at ESRF:*
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Names and affiliations of applicants (* indicates experimentalists):

Per-Anders Carlsson*, Peter Velin*, Andreas Schaefer* and Felix Hemmingsson*
Department of Chemistry and Chemical Engineering and Competence Centre for Catalysis, Chalmers University of Technology, 412 96 Gothenburg, Sweden.

Johan Gustafson*, Helen Edström* and Kim von Allmen*
Department of Physics, Lund University, 221 00 Lund, Sweden.

Report:

This report concerns a proposal aiming at, on the one hand, synchronous energy dispersive X-ray absorption spectroscopy (ED-XAS) and diffuse reflectance infrared Fourier transformed spectroscopy (DRIFTS) and, on the other hand, high-energy X-ray diffraction (HE-XRD) in an *operando* approach as to study supported Pd-Au model catalysts fosucing on i) the alloy/core-shell transition dynamics during redox cycling and ii) the catalyst structure determining the activation of methane (CH_4) for total oxidation. During the beamtimes at both ID24 and ID15, modulation excitation spectroscopic and scattering measurements were conducted fully exploit the potential of the ID24 and ID15A beam lines and extract catalyst structure-function relationships based on time-resolved measurements.

Due to several experimental challenges, the Pd-Au model catalyst was difficult to measure at ID24. Some data were gathered but it is challenging to make a proper analysis. Even if time has passed, we will revisit this data again later on. At ID15 the Pd-Au catalysts were measured as were also Pd-Pt and pure Pd catalysts. Unfortunately, the analysis of these results was interrupted due to redirection of PhD project. Also here, however, the data will be revisited in the future.

Due to these challenges at ID24, however, we also conducted experiments over a series of alumina and zeolite supported Pd catalysts focusing on the effect of water on the lean methane oxidation. These measurements were successful and were complemented with infrared experiments and transmission electron microscopy characterization in our home laboratories. In summary we show that water is shown to hamper the redox dynamics of realistic palladium oxide nanoparticles dispersed onto alumina and hydrophobic zeolite supports thereby lowering the activity for total oxidation of methane. The adsorption of water results in hydroxyl ad-species that block the methane and oxygen dissociation, and also seem to prevent lattice oxygen to be reactive species in the methane oxidation. The main catalytic action is thus shifted from the Mars-van Krevelen type of mechanism under dry conditions to a slower route with Langmuir-Hinshelwood type of steps in wet conditions. This is a key finding that has clear impact on how catalysts should be designed for low-temperature methane combustion for emission control.

The results have been published in a communication article [1] and a dissertation [2]

[1] P. Velin *et al.* in *ChemCatChem* 13 (2021) 3765-3771.

[2] P. Velin, *Spectroscopic investigation of palladium catalysts during wet methane oxidation,* dissertation at Chalmers University of Technology, 2020.