EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

ESRF

Experiment Report Form

ESRF	**Experiment title:** **Novel highly active iron based catalysts for carbon monoxide oxidation**	**Experiment number**: CH 5403
Beamline:	**Date of experiment**: from: 08 June 2018 to: 12 June 2018	**Date of report**: 20.08.2018
Shifts:	**Local contact(s)**: Dipanjan Banerjee, Alessandro Longo	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Prof. Dr. Matthias Bauer, Universität Paderborn, Warburger Str. 100, 33098 Paderborn

* Steffen Schlicher, Universität Paderborn, Warburger Str. 100, 33098 Paderborn
* Roland Schoch, Universität Paderborn, Warburger Str. 100, 33098 Paderborn
* Marina Huber, Universität Paderborn, Warburger Str. 100, 33098 Paderborn
* Jakob Steube, Universität Paderborn, Warburger Str. 100, 33098 Paderborn

Report:

Introduction: Iron oxide based catalysts on alumina support show promising results in carbon monoxide oxidation experiments. To increase their activity to the level of noble metal catalysts, they have to be optimized by means of their structural properties. Thus, it is crucial to draw structure-activity correlations and to understand, how variation of different parameters in synthesis of a catalyst influences its structure and as a result its catalytic performance. In the herein proposed experiments we mainly wanted to carry out X-ray absorption spectroscopy of the catalysts during CO oxidation experiments. This will give insights into the mechanism of catalytic carbon monoxide conversion on iron oxide nanoparticles, as this is still discussed controversially.

Experimental: During the beamtime at BM26A operando XANES and EXAFS spectra at the Fe K-edge of two different ironoxide@alumina catalysts in a defined gas stream containing 1000 ppm CO and 10 vol% oxygen in argon/helium at 300 ml/min were measured. For these experiments our self-designed operando reactor for X-ray absorption spectroscopy (XAS) was used in fluorescence mode (figure 1) which provides the possibility to measure spectra under conditions similar to the reaction conditions used in our home laboratory. Therefore catalysts prepared by incipient wetness impregnation of gamma-alumina with 2.5 wt% and with 10 wt% iron loading were chosen. Both catalysts were investigated ex-situ as solid samples (pressed to a pellet, 13 mm radius, 1 mm thick, diluted in BN) as reference, as well as in-situ in the above mentioned gas stream. With each catalyst one experiment with continuous heating from room temperature up to 600 °C for pretreatment as well as measurements at the temperatures corresponding to 25 %, 50 %, 75 % and 100 % yield were carried out. Besides the operando experiments and especially during the cooling of the cell XAS spectra of other catalysts with different loadings and different preparation techniques were measured, to compare them to the in-situ catalysts.

Figure 1: operando XAS reactor installed at BM26A, before alignment of the detector.

Results: Typical examples of the obtained spectra are shown in figure 2. It can be seen that there are quite distinct differences in the XANES region at different stages of the treatment and the actual reaction, but the data obtained are still under evaluation, especially the interpretation of the EXAFS region.

Conclusion: The obtained spectra show clear differences during pretreatment and the actual reaction which will lead to a better understanding of the following questions, which will be adressed in a publication in a high impact journal:

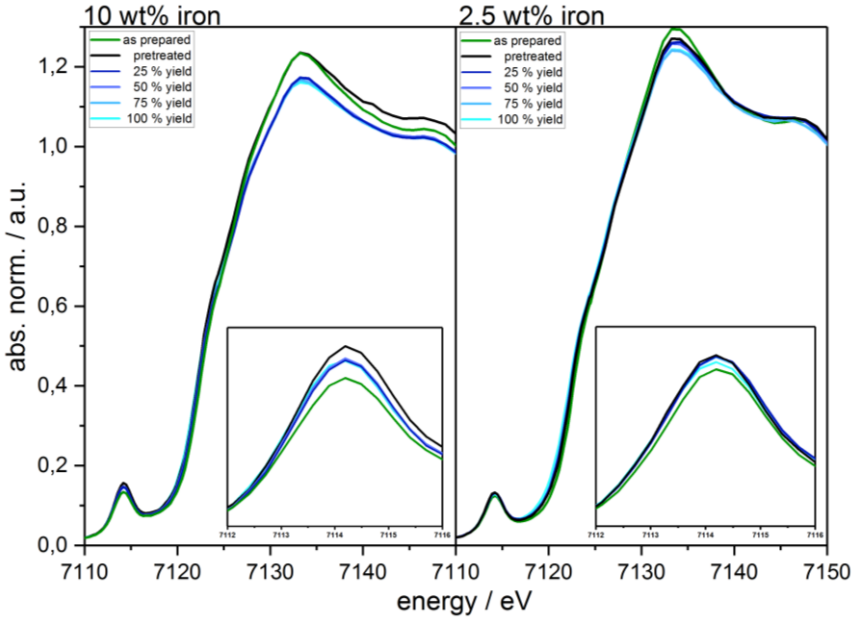

Figure 2: Prepeak and whiteline regions of the tested catalysts, each before and after pretreatment and at different stages of the carbon monoxide oxidation experiment.

- **How does pretreatment alter the geometric structure of iron species?**
- **How does the coordination geometry change during CO oxidation reaction?**
- **Which particle size and coordination geometry are benefitial for good activities?**
- **What is the actual mechanism of this reaction?**