DUTCH-BELGIAN BEAMLINE AT ESRF

EUROPEAN SYNCHROTRON RADIATION FACILITY

Experiment Report Form

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

(next page)

DUBBLE	**Experiment title:** XANES and EXAFS study of the activation and deactivation of Fe-based Fischer-Tropsch Catalysts	**Experiment number**: **26-01-779**
Beamline: BM26A	**Date(s) of experiment**: From: 25-04-2007 To: 30-04-2007	**Date of report**: 2 - 10 - 2007
Shifts: 15	**Local contact(s)**: Dr Sergey NIKITENKO (e-mail: nikitenko@esrf.fr)	

Names and affiliations of applicants (* indicates experimentalists):

Emiel de Smit, dr. Frank de Groot, Prof.dr. Bert Weckhuysen, Department of Inorganic Chemistry and Catalysis, Utrecht University, The Netherlands

Experimentalists

Emiel de Smit*, Dr. Andy Beale*, Elena Sacaliuc*, Ad van der Eerden*, Department of Inorganic Chemistry and Catalysis, Utrecht University, The Netherlands

Report: (max. 2 pages)

During this beamtime, work was focused on gaining insight into the iron phases responsible for the activation and deactivation of Fe-based Fischer-Tropsch (FT) catalysts. The largest part of the time was spent on collecting *in situ* data during the activation and reaction of the catalysts. The remainder of the time was spent on *ex situ* analysis of reference materials and spent catalysts.

XANES/EXAFS/WAXS study of the activation of Fe-based Fischer-Tropsch catalysts

In preparation of the beamtime it was decided to include the WAXS facilities that beamline 26A has in the research. As Fe-based FT catalysts show changes in bulk composition during activation and reaction, using WAXS in combination with XAS can provide important complementary information on the bulk state of the catalysts.

Three different Fe-based catalysts; bulk α-Fe_2O_3, Fe_2O_3/CuO/K_2O/ZnO and Fe_2O_3/CuO/K_2O/SiO_2, were studied under identical conditions. The catalysts were heated to 250^oC in a mixture of 5% H_2/He and 5% CO/He, while WAXS and XANES data were recorded continuously. During reaction, reaction products were monitored using a mass spectrometer. After different FT reaction times, EXAFS data was collected in the quick EXAFS mode.

Unfortunately, the catalyst systems behaved different than expected and only very small changes were observed in the WAXS and XAS data, even after prolonged reaction times (~4 h) (Figure 1). As the FT reaction rate is first order in the partial pressure of H_2, this was most probably the result of the use of dilute gases. The analysis of reaction products, however, showed that there was still some catalytic activity of the samples, giving a strong indication that bulk species are not necessary for the catalysts to be active in the FT reaction.

Overall, the quality of the recorded XAS and WAXS data was very good and new insights were gained into the catalytic system. The project will be continued in the beamtime in the fall of 2007 (proposal 26-01-788), applying the new insights and experience gained from this run.

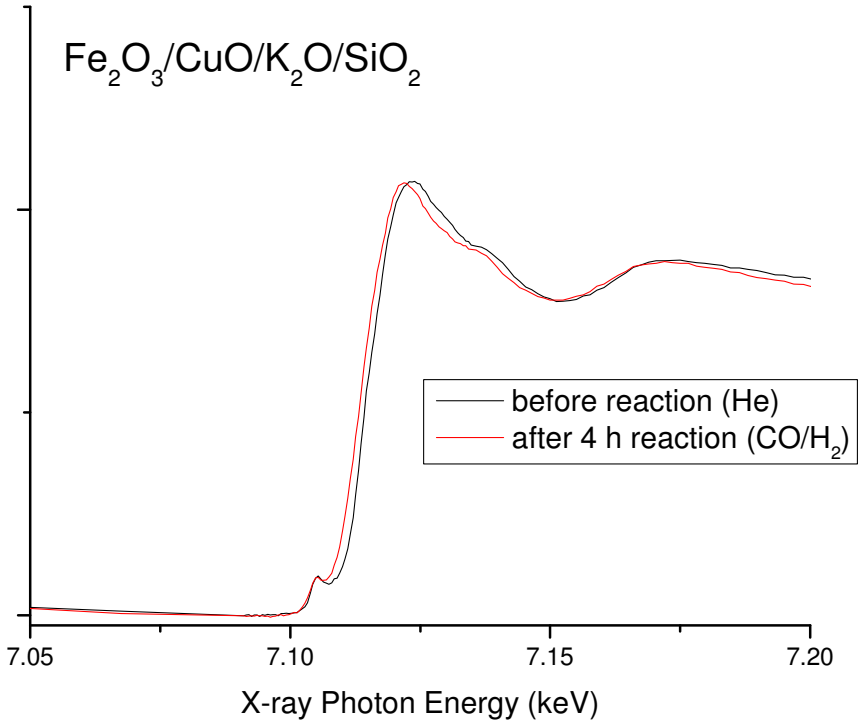

Figure 1: XANES spectra of the Fe_2O_3/CuO/K_2O/SiO_2 catalyst before reaction (black) and after 4h of reaction time (red)

***Ex situ* analysis of reference materials and spent catalysts**

WAXS/XANES/EXAFS data was recorded on Fe_2O_3, Fe_3O_4, Fe_xC and Fe reference materials, as well as on some previously used catalyst samples. The data quality was excellent and therefore it is to be expected that some of these results will be used in future publications.

Conclusions

Although the experiments did not yield the expected results, important new insights were gained into the catalytic system. Overall, the user support on the beamline was outstanding and we are confident that the planned future experiments will be successful.