EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office via the User Portal: *https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do*

Deadlines for submission of Experimental Reports

Experimental reports must be submitted within the period of 3 months after the end of the experiment.

Experiment Report supporting a new proposal ("relevant report")

If you are submitting a proposal for a new project, or to continue a project for which you have previously been allocated beam time, you must submit a report on each of your previous measurement(s):

- even on those carried out close to the proposal submission deadline (it can be a *"preliminary report")*,
- even for experiments whose scientific area is different form the scientific area of the new proposal,
- carried out on CRG beamlines.

You must then register the report(s) as "relevant report(s)" in the new application form for beam time.

Deadlines for submitting a report supporting a new proposal

- 1st March Proposal Round - **5th March**
- 10th September Proposal Round - **13th September**

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

Reports on experiments relating to long term projects

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report in English.
- include the experiment number 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.

ESRF	**Experiment title:** In situ Tomographic Imaging of a Silica-Supported Zirconocene-Based Catalyst Particle Using Holotomography	**Experiment number**: CH-6155
Beamline:	**Date of experiment**: from: 18.11.21 to: 23.11.21	**Date of report**: 16.12.2021
Shifts:	**Local contact(s)**: VILLANOVA Julie, VANPEENE Victor	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Maximilian Werny, Rafael Mayorga-González, Wilhelmina Carolina Versluis:

Utrecht University

Inorganic Chemistry and Catalysis group

Luca Carnevale:

University of Twente

BIOS lab-on-a-chip group

Report:

Our user group arrived two days in advance to discuss operations with the beamline scientists (Julie Villanova, Victor Vanpeene) and the head of the chemistry laboratory (Harald Müller). Parts of the experimental set-up were installed a day before the beam time. The first 6 h of the beam time were spent on finalizing the experimental set-up and leak-proofing all connections. The set-up was then checked and approved by the safety group.

Our first measurememements involved the *ex situ* characterization of individual catalyst particles in the chip-based microreactor system at 17.4 keV (tomographies recorded over 360 degrees). Based on the preliminary reconstructions, we were able to conclude that high resolution tomographies can be recorded with the chip-based reactor (Si_3N_4 capillary allowed for high X-ray transmission, no or very limited sample movement in the capillary).

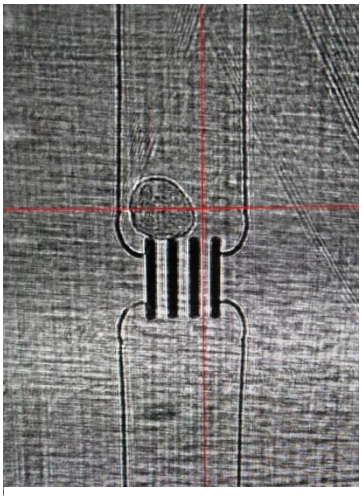

Figure 1. Catalyst particle loaded inside the chip-based micro-reactor system with slits.

The reactor was then connected to the remaining set-up (gas lines, MFCs, back pressure regulator) and pressurized. Unfortunately, we did not observe a pressure build-up with nitrogen. Upon closer inspection of this first reactor, we located a small hole in the capillary of the reactor (before particle trap). The reactor was most likely damaged during loading with the first particles. Due to this leak, we could not proceed with the ethylene polymerization experiment.

The procedure was repeated with two additional reactors that were loaded with individual catalyst particles. Both reactors leaked signficantly at the interface between the chipholder and the chip. The etching of the chips during fabrication

most likely reduced their thickness, thus leading to a suboptimal contact with the chipholder. Tests performed on previous batches of these reactors did not reveal such anomalies in thickness. We therefore did not expect these issues with the new batch of reactors that was brought to the beamline.

Our final reactor was damaged while loading it with catalyst particles. The part of the capillary after the trap was ruptured.

The remaining beam time was dedicated to the *ex situ* characterizaion of different catalyst systems (silica-supported zirconocene catalyst, Fluid Catalytic Cracking catalyst) in their pristine state as well as in the presence of carbon-based products (e.g., polyethylene, polylpropoylene, coke). For selected samples, we measured individual particles before and after removal of the carbon-based phase. By aligning the two tomographies and computing their difference, we are aiming to locate the carbon-based phase in 3D.

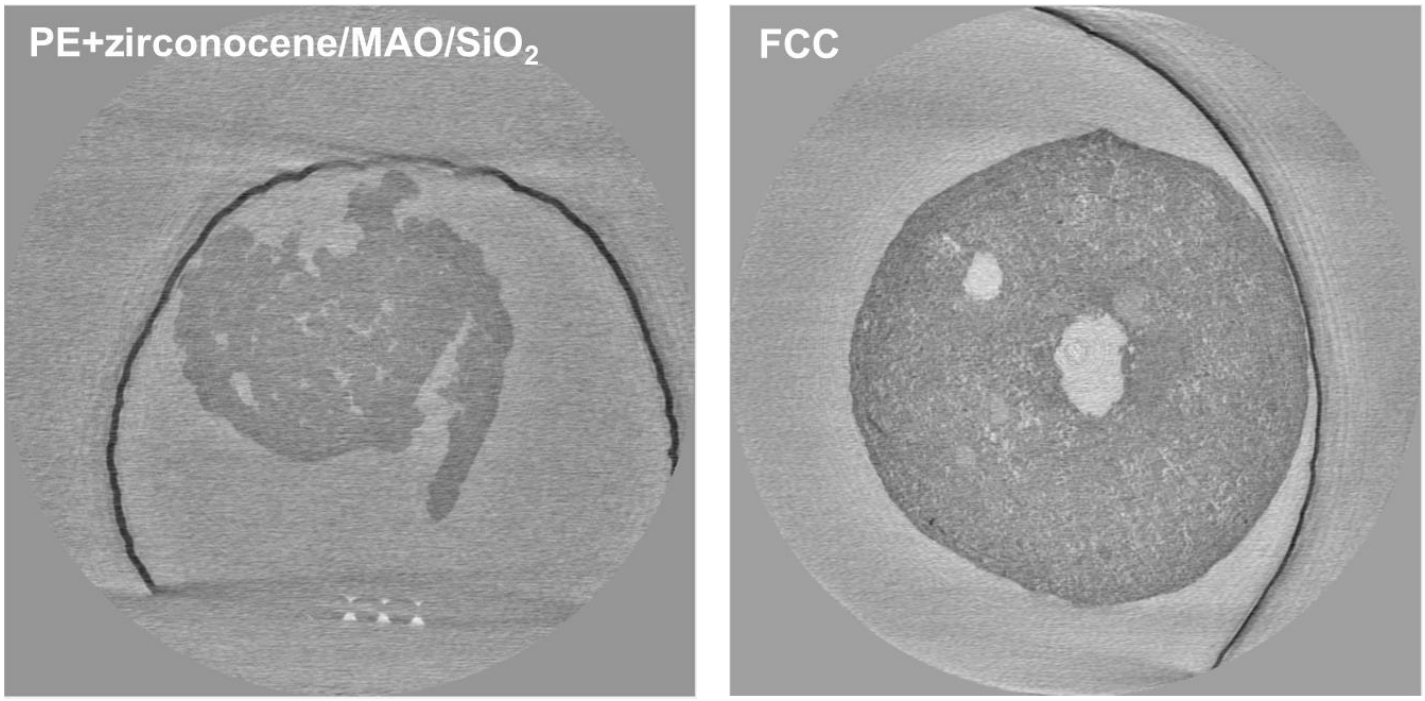

Figure 2. Virtual slices (xy) of a pre-polymerized zirconocene-based catalyst particle (polyethylene/SiO_2 composite material, PE phase is dominant) (left) and a pristine fluid catalytic cracking catalyst particle (composite of silica, alumina, zeolite, binder) (right). Both tomographies were acquired at 17.4 keV.

Future efforts will be directed towards securing a larger number of functioning reactors for any potential beam time. The fabrication of airtight reactors represents a complex procedure that still requires additional fine-tuning. The reactor technology is, however, ready for application under high temperature (300 °C), high pressure (up to 20 bar) reaction conditions, as is clear from our in-house temperature and pressure testing.