ESRF	**Experiment title:** Investigation of pore structures of sol-gel synthesized Ni-based methanation catalysts by PXCT to study the influence of structure directing agents	**Experiment number**: CH-5805
Beamline: ID16a	**Date of experiment**: from: 02/12/2020 to: 06/12/2020	**Date of report**: 27/02/2021
Shifts: 9	**Local contact(s)**: Peter Cloetens and Federico Monaco	*Received at ESRF:*

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Report:

Please note this is a preliminary report - data processing of all results is not yet complete at the time of reporting. The experiments of this report were carried out via the mail-in and remote procedure. The experiments were carried out at ID16A of ESRF by Federico Monaco and Peter Cloetens. 7 different samples were shipped to ESRF already mounted on aluminum tomography pins by focused-ion beam preparation. All samples were Ni-based methanation catalysts on different support materials, alumina (4 samples) and zirconia (3 samples). The support materials of the catalysts are characterized by different pore strucutres depending on their synthesis and thus exhiting different mass transport properties, which is important for the catalytic performance.

Holographic tomography and near-field ptychographic tomography were applied to study the pore structures of the different materials. To confirm the presence of Ni and its homogenoeus distribution in the samples, a X-ray fluorescence (XRF) map was acquired before the actual tomography experiments. The general holographic tomography scans were acquired with a field of view covering the whole particle (diameters of all samples about 50 to 60 µm) with a pixel size of 25 nm. Additionally, region of interest tomography with a pixel size of 7 nm and smaller field-of-view was performed after the general holographic tomography scans.

In previous TEM-EDX mapping of the $NiFe/Al_2O_3$ sample always a homogenoeus distribution was found, however the 2D XRF

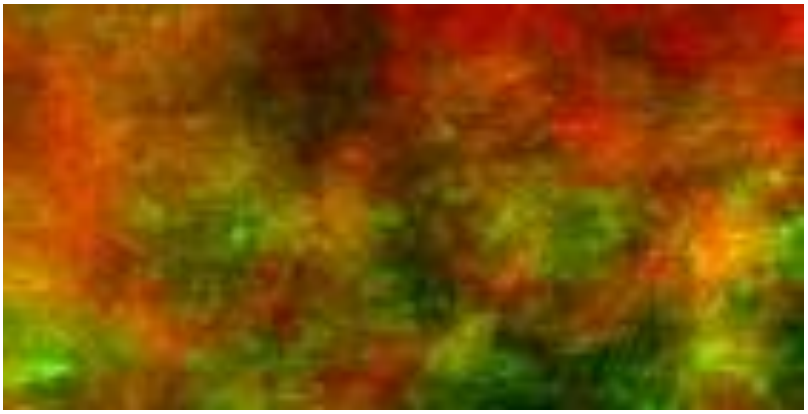

Figure 1 2D XRF map of the $NiFe/Al_2O_3$ catalyst, Ni red, Fe green

map before the experiments showed a quite heterogenous distribution of Ni and Fe (step size of 200 nm). The pixel size here is as large as the typical field of view of TEM-EDX mapping, and thus allows to uncover heterogenity of the material on longer length scales as shown in Figure 1. This suggests that TEM-EDX did not have sufficient field of view to address the heterogeneity of the sample Thus, additionally a XRF tomography slice was acquired of the $NiFe/Al_2O_3$ catlalyst to analyze the spatial distribution of Ni and Fe in the catalyst particle.

To obtain information about the electron density and therefore examine the presence of pores, the Ni/Al_2O_3 and $NiFe/Al_2O_3$ catalysts were also studied by near-field ptychographic tomography. Thus, porosity information below the resolution limit of the resulting tomograms can be indicated by later electron density analysis. The reconstruction of all tomograms is not finished yet, thus also the subsequent image analysis is only briefly reported. For the Ni/Al_2O_3 catalyst, the porosity analysis results obtained from image analysis of the reconstructed volume from holographic tomography with 25 nm pixel size is shown in Figure 2. One can observe a relatively uniform distribution of the pores around 1.5 µm, while some large pores are additionally found. The porosity information can be applied by our collaborators for macrokinetic calculations and thus simulation of the performance of the material.

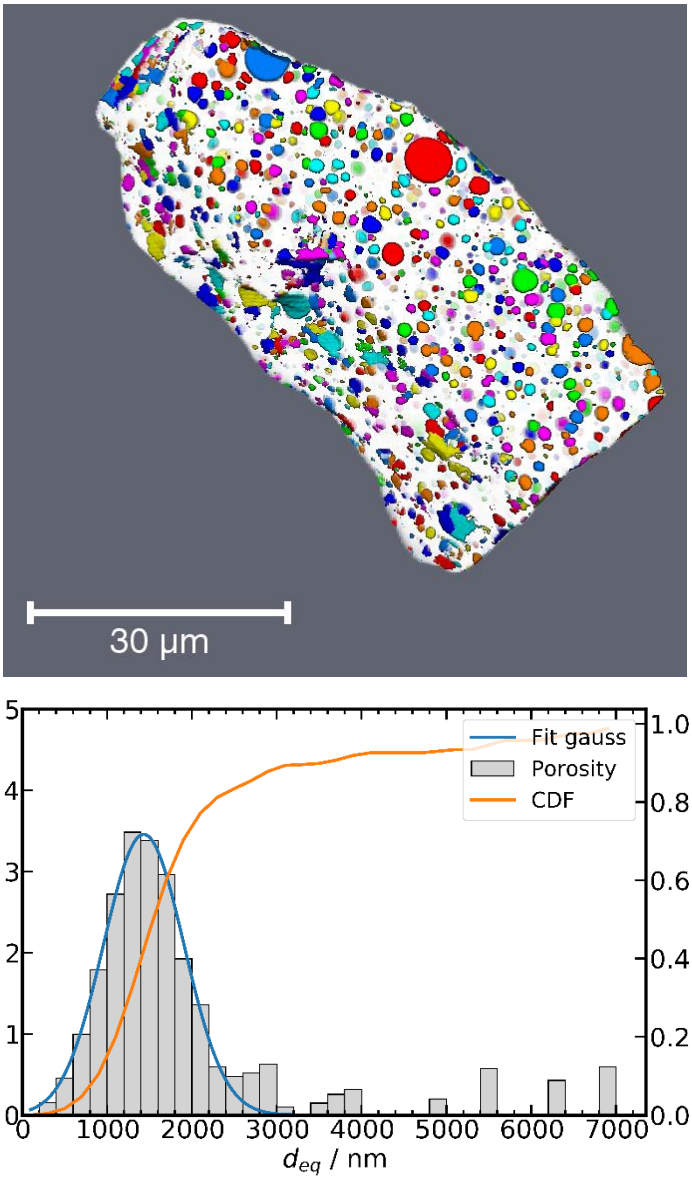

Figure 2 Labelled pores after image anaylsis of the reconstructed 25 nm holographic tomogram of the Ni/Al_2O_3 sample (top); Porosity distribution of the obtained from the quantitative image analysis (bottom).

No comparison of the influence of the different support materials could be done so far due to the ongoing data processing and data analysis. However, from an initial justification of the already received data, we conclude that the experiment was successful and a future publication is planned.