ESRF	**Experiment title:** **Green steel: The evolution of porosity during hydrogen-based direct reduction of iron oxide**	**Experiment number**: MA-4880
Beamline: ID16B	**Date of experiment**: from: 15 September 2021 to: 18 September 2021	**Date of report**: 20 February 2022
Shifts: 9	**Local contact(s)**: Dr. Julie Villanova	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Dr. Yan Ma*, Max-Planck-Institut für Eisenforschung, Germany

Prof. Dr. Guillermo Requena, Deutsches Zentrum für Luft- und Raumfahrt (DLR) and RWTH Aachen University, Germany

Report:

Fossil-fuel-free ironmaking is indispensable for reducing massive anthropogenic CO_2 emissions in the steel industry. Hydrogen-based direct reduction (HyDR) is among the most attractive solutions for green ironmaking, with high technology readiness. In HyDR, pelletized solid iron ores (hematite and magnetite) are reduced by hydrogen gas. Reaction kinetics is affected by micro-to-atomic-scale features of the different oxides and the adjacent iron layers, including crystal defects, porosity, mechanics, and local composition [1-3]. Particularly, porosity is a critical factor when gaseous diffusion and local storage/transport of water are rate-limiting steps in HyDR [4, 5]. The porosity and its tortuosity affect gas percolation in and out of the internal free surface regions (*i.e.*, inbound hydrogen and outbound water transport). To further understand the influence of the porosity on the reaction kinetics, time-resolved 3D quantification of the porosity (in terms of size, distribution, morphology, and connectivity) and the associated phase evolution (here, hematite (Fe_2O_3), magnetite (Fe_3O_4), wüstite (FeO), and reduced iron (Fe)) is absolutely necessary.

In this study, we performed *in-situ* high-resolution X-ray computed tomography (CT) enhanced by phase contrast to characterize the porosity and the adjacent phases at the beamline ID16B during the direct reduction of iron oxide with pure hydrogen. The needle-shaped specimens (with a diameter of ~100 µm) were prepared from direct-reduction pellets and single-crystal hematite, then they were encapsulated in a hydrogen atmosphere with quartz capillaries. The high-temperature reduction experiments were conducted by heating up the encapsulated specimens using a microheater at the beamline to 700 °C and 900 °C. The spatial resolution of the CT experiments was set as 30 nm by adjusting the sample-to-detector distance. The microstructure of the initial hematite pellet is shown in **Figure 1**a. Tne dark black regions represent the initial pores inherited from the pelletization process, while the bright grey regions are hematite grains. The *in-situ* high-resolution CT experiments were successful to visualize the porosity evolution. During HyDR, numerous pores were generated, as revealed in **Figure 1**b (in the similar region in **Figure 1**a). Moreover, a hierarchical pore structure was observed in the hematite particles. A large number of fine pores formed at the edges of the hematite particles, while relatively larger pores can be found inside the hematite particles. The evolution of the pores in the same

region of the specimen (after registration processing) will be further analyzed in detail, in terms of their morphology, size distribution, spatial distribution, and connectivity in a 3D volume.

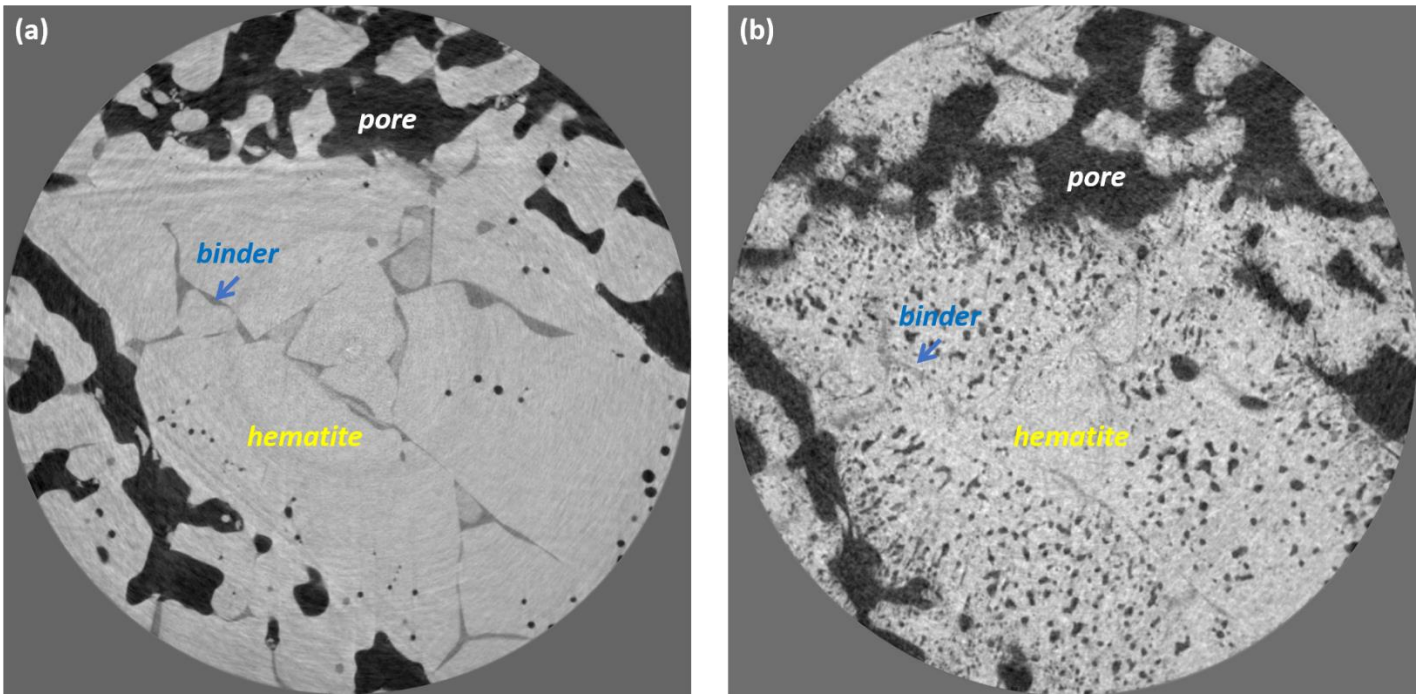

Figure 1 *Synchrotron X-ray computed-tomography (CT) slices of direct-reduction pellet: (a) initial state and (b) reduced state after reduction at 700 °C for 2 hours at the beamtime during in-situ CT experiments. The dark black regions represent pores.*

With further data analysis, it is very promising to reveal the porosity evolution in 3D during the direct reduction with hydrogen. The aim of the proposed experiment can be successfully achieved. This work will contribute to the new insights into the porosity evolution (in terms of its 3D distribution, size, morphology, and connectivity) during the gaseous reduction of iron oxides. In combination with our other results (*e.g.*, X-ray diffraction, electron backscatter diffraction, and phase-field simulation), a publication is in preparation. We expect the ESRF beamline scientist at ID16B to be as co-author of our publication.

Reference:

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