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Electron and X-ray Tomography of Iron/Iron Oxide Redox Reactions for Large-Scale Hydrogen Storage

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One of the most urgent problems that have to be solved in the near future is the energy problem and the environmental impact resulting from fossil fuels. For the transition to an environmentally clean energy production based on renewable sources like solar, wind and water power, it is evident that hydrogen will become one of the dominating energy carriers. An important aspect of hydrogen technology is its safe and economic storage. So far the storage as high-pressure gas is widely used, but the volumetric hydrogen capacity remains low and the high pressure is a safety concern. Other storage approaches are under investigation and relay on solid-gas reactions, like metal hydrides and highly porous materials, e. g. metal organic frameworks and carbon nanotubes.

The hydrogen storage method pursued in this study is based on the cyclic implementation of the redox reaction 3 Fe + 4 $H_2O \leftrightarrows Fe_3O_4$ + 4 H_2, Figure 1. This reaction is also known as steam-iron-process historically. Hydrogen generation results from the reaction of highly reactive nano-sized iron powder with water steam, and hydrogen storage is performed by the reaction of magnetite powder with hydrogen. A known disadvantage is the decrease of reaction turmnover (storage capacity) after repeated reaction cycles. The loss of storage capacity is caused by high temperatures that cause a particle coarsening and that result in a deactivation of the powder. Metal oxide additives in the reduced iron oxide can preserve the reactivity of the Fe/Fe_3O_4 powder during enduring cycling.

In this study we use electron and X-ray microscopy to characterize the morphology of such iron/iron oxide particles and of particle agglomerates before and after cyclic hydrogen storage. The application of both microscopic techniques is a challenging task for this material system. The size of the powder particles is in the range of few 10 nm to some 100 nm. The lab-based X-ray microscope (Xradia nanoXCT-100) allows investigations on powder particle agglomerates in the size of 10 µm. As there is no vacuum requirement, powder particle agglomerates can be imaged under atmospheric pressure and at elevated temperatures, see Figure 2a. We designed a reaction chamber that fits into the beam path of the X-ray microscope. Using this attachment, the morphology change of powder particle agglomerates during the half reaction of the steam iron process can be imaged directly. Preliminary results from in-situ X-ray radiography and tomography tests inside a micro reactor chamber will be shown. The spatial resolution of the X-ray microscope is in the range of the particle size, which makes it difficult to observe the smallest particles and details at the particle interaction sites. On the other hand, the number of particles observed at once in a TEM (ZEISS Libra 200 Cs MC) is restricted by the absorption length of electrons, especially in tilt series for tomographic reconstruction, see Figure 2b. The combination of results from both, TEM and X-ray microscopy, enhances the interpretation of the materials under investigation.

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It will be shown that an optimized powder composition as well as a pretreatment of the iron oxide powder by ball-milling leads to an improvement of the capacity after repeated storage cycles.

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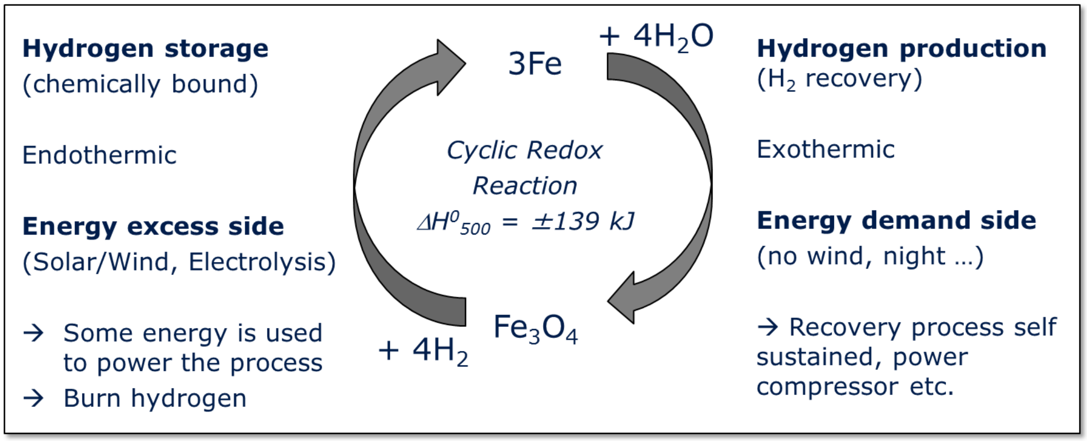

Figure 1. Scheme of the steam-iron-process as cyclic driven redox reaction for hydrogen storage

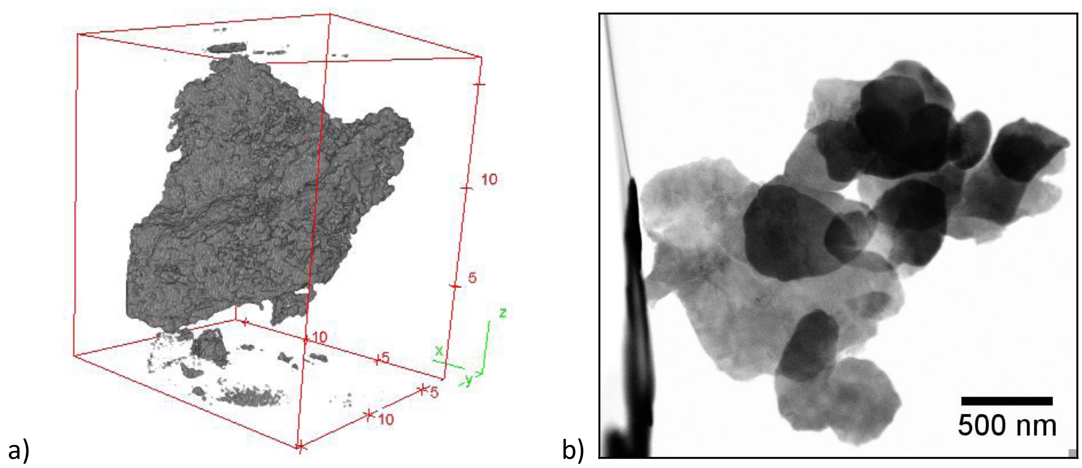

Figure 2. Iron particles after reduction in hydrogen atmosphere (a) reconstructed volume of a larger agglomerate after a reduction experiment inside the microscopic reaction chamber (scale in µm) (b) STEM dark field image (inverted) of some particles.

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