Iron oxide reduction in a high-performance microwave argon-hydrogen plasma

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Global steel production accounts for a substantial part of man-made CO_2 emissions. The utilization of green hydrogen in future industry processes has the potential to render iron ore reduction nearly climate-neutral [1, 2]. Microwave-sustained thermal hydrogen plasmas can be employed for iron oxide reduction, with the end products being H_2O and Fe. For instance, the advantages are faster reduction rates and lower energy consumption than existing methods. Using plasmas also enables the production of various reactive species like atomic hydrogen, hydrogen ions, and excited H_2 molecules. The effects of especially vibrationally-excited H_2 molecules and hydrogen ions on the reduction process at the interface between the plasma and an iron/iron oxide surface have already been investigated in a global equilibrium model [3]. Further experiments focusing on microscopic process parameters and non-equilibrium surfaces processed during the reduction are not sufficiently addressed. This study is intended to follow up on these research questions and to investigate the interface between iron oxide and hydrogen.

Therefore, an argon-hydrogen microwave plasma is used for iron ore reduction by either injecting an iron oxide powder into the gas flow or exposing defined solid samples to the plasma. The samples primarily consist of hematite (Fe_2O_3). The main reaction equation is then:

$$Fe_2O_3 + 3H_2 \longrightarrow 2Fe + 3H_2O.$$

The plasma is generated in a resonator and is sustained by a 6 kW-capable microwave source. The combination of an axial as well as two tangential gas inflows generates a swirl-like gas flow pattern, which stabilizes the plasma on the central axis and ensures steep radial temperature gradients to protect the wall of the discharge tube. Furthermore, this swirl flow shall prevent the iron oxide particles from adhering to the tube's surface. Without the presence of the swirl flow, the plasma would burn directly on the surface of the tube. Below the resonator, the plasma subsequently enters a water-cooled double-walled reactor chamber where solid samples can be treated on a substrate holder by the plasma-produced reactive species. Alternatively, iron oxide particles can be added to the axial gas flow. A specially designed particle feeder is employed to ensure defined admixtures. Fig. 1 shows a schematic of the experimental setup for the two different operation modes.

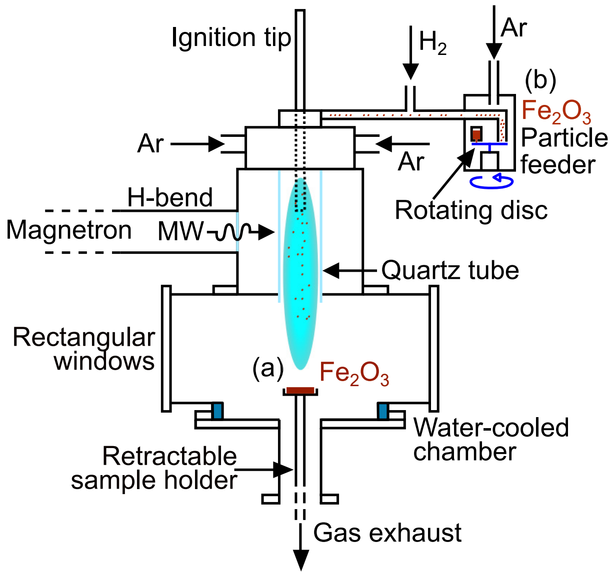

Fig. 1: Schematic of the experimental setup of the microwave discharge for the two operation modes: (a) – using a defined solid sample of hematite (Fe_2O_3) and (b) – injecting a defined amount of hematite particles into the gas flow.

A design study is performed to view the motion, residence time, and location of melting/evaporation of injected particles in the presence of the swirl flow for the microwave discharge compared with a cylindrical ICP discharge. Particle size, gas flow, and temperature profiles are being varied. The degree of particle addition into the gas flow continuously transforms the plasma into a metal plasma, making optical diagnostics, especially laser diagnostics, rather challenging due to the strong scattering background.

Thus, for in-situ investigations, optical emission spectroscopy is used to examine the influence of metal particles on the properties of the plasma. Furthermore, a major focus lies in determining the gas temperature, as it is a crucial parameter for both analytical considerations and simulations of physical or chemical reactions. As ex-situ diagnostics, XRD depth profiling, XPS, and SEM are employed to characterize the interaction surface between the hematite sample and the hydrogen and quantify the degree of reduction.

A similar setup using a high-performance microwave discharge is already operated for hydrogen production via plasma methane pyrolysis [4]. There, optical emission spectroscopy has been conducted to measure the emission of black body radiation from hot carbon particles and of the dicarbon Swan bands. The evaluation revealed a gas temperature of around $4.500\,\mathrm{K}$ in the center below the resonator, as seen in Fig. 2, where spatial-resolved profiles of the gas temperature are depicted [5].

In conclusion, this study investigates the microscopic interface between iron oxide samples and reactive hydrogen species produced by a high-performance microwave plasma by applying spectroscopic and material-characterizing diagnostics. Thereby, the treatment of solid samples as well as of powders of hematite is considered. A major focus lies in determining the gas temperature as a crucial variable for the iron ore reduction process.

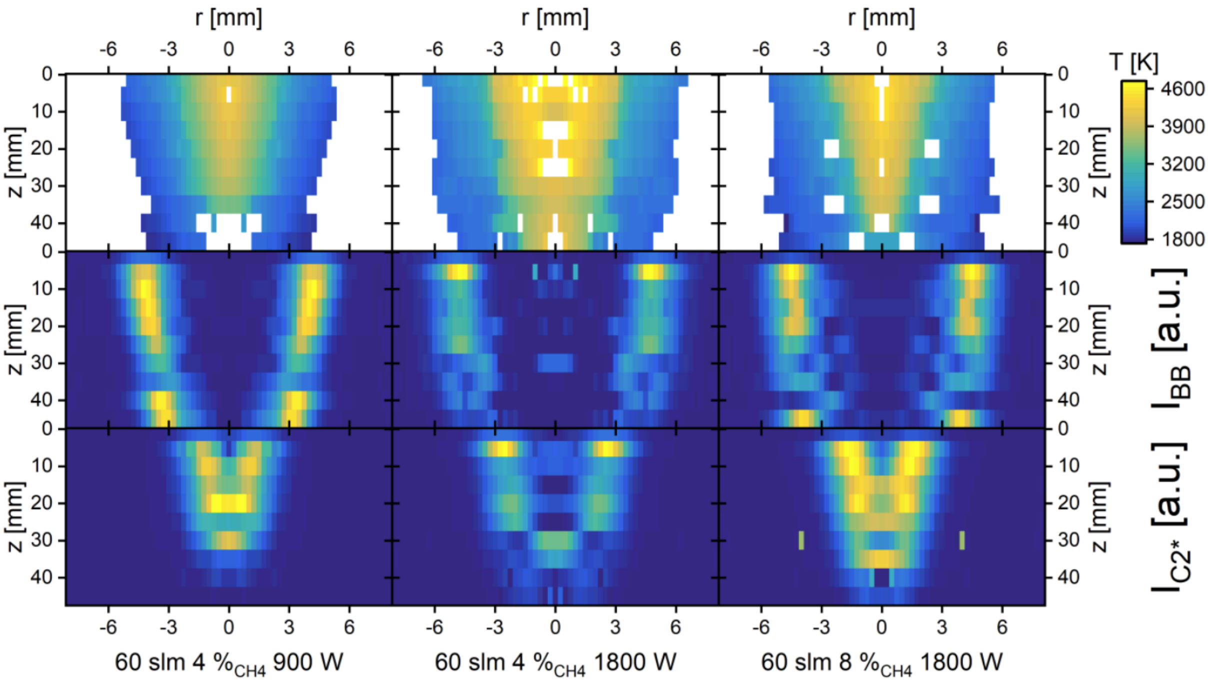

Fig. 2: Space resolved profiles of the gas temperature (T), the emission of the black body radiation (I_{BB}) and the emission of the dicarbon Swan bands (I_{C2^*}) as a function of the radial and axial position for three different sets of operating parameters in an Ar/CH_4 microwave plasma. Retrieved from [5].

Acknowledgment

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