

Combustion of Rechargeable Metal Fuels in Fluidized Bed Boilers

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Table of contents

Table of contents.....	2
1 Introduction.....	3
2 Methodology	4
2.1 Introduction of the materials	4
2.2 Material characterization	6
2.2.1 Minimum fluidisation velocity	6
2.2.2 Oxidation degree	7
2.2.3 Solid density.....	8
2.2.4 Particle size distribution	9
2.3 Fluidisation regime.....	11
3 Experimental description	14
3.1 Setup of the test rig.....	14
3.2 Data recording	17
3.3 Description of the test campaign	19
3.4 Occurred limitation of experimental operation	20
4 Results and discussion	22
4.1 Assessment of iron powders.....	22
4.2 Results of the reduction experiments	25
4.2.1 Iron ore powder	25
4.2.2 Iron powder	27
4.3 Results of the oxidation experiments	30
5 Conclusion	34
5.1 Selection of iron powder	34
5.2 Determined window of stable operation.....	34
6 References.....	35
7 Appendix.....	36

1 Introduction

The overall aim of the CORAL project is to support the transition from fossil fuels to renewable energy by developing an alternative, CO₂-free energy storage system using iron as fuel. In this sense, it is being investigated how existing coal-fired power plants can be retrofitted to enable the combustion of iron powders and the associated storage of energy. During the project, the process will be conducted at three different scales: a lab-scale fluidised bed, a medium-scale fluidised bed, and a 1 MW pilot plant.

The aim of the small-scale fluidised bed tests is to determine a window of stable operation, as well as to attain a level of knowledge for the well-founded selection of a suitable metal powder. The specific objectives of the lab-scale tests are:

- Determination of the minimum fluidisation velocity
- Determination of the window of stable operation
- Investigation of the effect of different temperatures on the iron conversion and the hydrodynamic stability.
- Investigation of three different iron powders to determine the most suitable powder for the following experiments

These objectives were set to obtain the most suitable material for energy storage with the best reaction properties, thereby enabling further investigations. The investigations into the influence of temperature and gas composition lead to a better understanding of the behaviour of oxidation and reduction of iron powder in a fluidised bed. Establishing a window of stable operation is also a fundamental prerequisite for successfully conducting further investigations and guaranteeing stable operation.

Following this introduction, the report includes a description of the lab-scale experimental setup. Afterwards, the three choices of metal powder are presented with their respective properties. Finally, the tests in the lab-scale fluidized bed are described and discussed.

2 Methodology

2.1 Introduction of the materials

Four iron powders from POMETON and an iron ore powder were used for the tests. The examined materials differ in their particle properties. Three of the iron powders are commercially available products from POMETON. These are Ferelet MT 425/3.0, Ferelet RI 425/3.7 and Feresint RI 250/2.1. Here RI stands for a further reduction after the production of the iron powder to obtain an increased degree of reduction. The powder labelled MT, on the other hand, was not subsequently reduced. The first number also indicates the maximum permissible particle size. The second number is the apparent density. In addition, an experimental powder was produced by POMETON, which has a slightly higher porosity due to the alternative manufacturing process (Iron Powder Type 120875). In addition, an iron ore from the Tierga mine in Spain was analysed in preliminary tests.

POMETON describes Ferelet MT 425/3.0, Ferelet RI 425/3.7 and the Iron Powder Type 120875 as follows:

Ferelet MT 425/3.0

This powder features a highly porous, irregular morphology with a relatively low oxygen content (0.52%). Its specific surface area supports high oxidation kinetics, making it an ideal candidate for research-oriented or batch-based combustion systems where fast ignition and reactivity are prioritized. However, its lower packing density and moderate flow properties may limit its effectiveness in continuous feed systems.

Ferelet RI 425/3.7

Designed for industrial use, this grade demonstrates excellent flowability (23 s/50 g) and the highest packing density (3.61 g/cm³). With the lowest oxygen content among the three (0.13–0.21%), it delivers superior thermochemical efficiency and process stability in multi-cycle combustion systems. Its dense, spherical particles make it particularly well-suited for fluidized bed reactors and scalable MEC systems

Water atomized Reduced Iron Powder 120875 (WRIP)

This powder offers a compromise between reactivity and flow properties. While it has a relatively low oxygen content (0.24%) and a moderate surface area (3.77 m²/kg), its low apparent density (1.71 g/cm³) and higher flow resistance (50 s/50 g) suggest some limitations in high-throughput environments. Its internal porosity and spherical shape make it applicable in systems where increased oxidation surface is desired, but dosing consistency must still be managed carefully.

Additionally, the powder Feresint RI 250/2.1 can be describes as:

Feresint RI 250/2.1

This powder consists of significantly smaller particles than the other powders, which leads to a higher surface area to mass ratio. The high surface area to mass ratio supports high reaction kinetics and makes it theoretically very suitable for achieving a high degree of reaction. However, this property also favours rapid agglomeration, making it difficult to test in a fluidized bed.

The examined materials were analysed for their particle properties prior to the experimental tests. The particle size distribution, solid density and degree of oxidation were determined for this purpose. The minimum fluidisation rate was also determined experimentally and estimated by calculation. These values are the starting point for the evaluation of the results presented in this paper. Table 1 shows the data of the materials before their use in the experiments. The iron ore (Tierga ore calcined) is fully oxidized, and the iron powders (Ferelet MT 425/3.0, Ferelet RI 425/3.7 and Feresint RI 250/2.1) are nearly completely reduced. From this, it can be concluded that the calculated data for reduction and oxidation degrees were correctly evaluated. The procedure for determining the respective values is given in section 2.2.2. The particle size distribution as described in 2.2.4 is shown in Figure 4.

Table 1 Initial properties of materials

Material	d_{10} in μm	d_{50} in μm	d_{90} in μm	d_{32} in μm	Solid density in kg/m^3	Bulk density kg/m^3	Oxidation degree in %
Feresint RI 250/2,1	28.28	75.37	152.67	64.2	7874	2090	0
Ferelet MT 425/3,0	119.52	181.06	255.68	172.5	7424	3088	2.07
Ferelet RI 425/3,7	119.40	177.77	281.52	177.9	7601	3716	0.21
Iron Powder type 120875	158.16	190.00	221.28	191.5	6795	1638	5.24
Tierga ore	191.51	235.61	178.10	232.3	4493	1847	100

2.2 Material characterization

2.2.1 Minimum fluidisation velocity

The minimum fluidisation velocity (u_{mf}) is determined by the drop of pressure over the material bed. For this purpose, the gas inflow is gradually increased, and the pressures are recorded via a measuring system. After exceeding the velocity at the loosening point of the fluidized bed, the fluidisation of the solid bed can be detected by a rise of bubbles in the reactor and the resulting movement of the surface. The fluid volume flow is further increased to map the area of constant pressure loss. The superficial velocity is then gradually reduced until the solid bed solidifies. The evaluation determines the superficial velocity for the individual steps, and the logarithm of the pressure difference is plotted over this. Figure 1 shows an example of the evaluation for determining the minimum fluidisation of the sample Tierga ore calcined (fractionated to $x > 212 \mu\text{m}$). The calculation of the minimum fluidisation velocity is carried out by applying two straight lines to the proportional section and the section of constant pressure difference and determining the point of intersection.

The increase in pressure to overcome the initial solidification is also clearly recognisable.

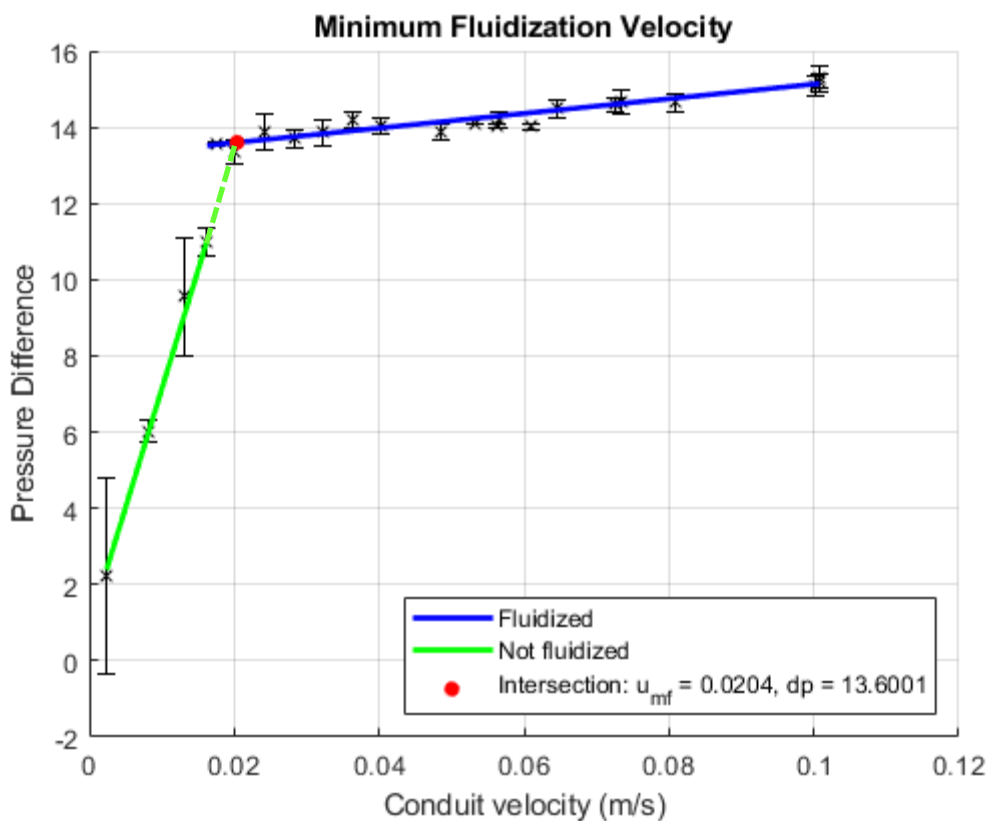


Figure 1 Minimum Fluidisation Velocity Feresint

In addition, the theoretical minimum fluidisation velocity is also calculated with the Wen–Yu–Correlation [2] using the previously determined PSD of the starting material for a temperature of 25 °C and 750 °C. The mean diameter (d_{50}) is used for the calculation. This provides an approximation of the behaviour of the real material and was used as a reference value. The

temperature of 750 °C was chosen as the reference value to simulate the conditions present during operation of the fluidised bed in the experiment. The minimum fluidisation velocity is used as a limit for the operating window.

Since the test rig used in the investigations in this paper can only be operated as a bubble fluidised bed, the minimum fluidisation velocity and discharge velocity set the limits for fluidisation.

Figure 1 shows an example of the evaluation of the minimum fluidisation velocity of the experimental investigations. The experimental investigations were carried out three times and the mean value of the results was calculated in order to obtain a representative result. In Table 2 the results determined for the four iron powders and the iron ore investigated.

Table 2 Fluidisation velocities

Material	u_{mf} exp. Tests in $\frac{m}{s}$ [20 °C]	u_{mf} calc. in $\frac{m}{s}$ [20 °C]	u_{mf} calc. in $\frac{m}{s}$ [750 °C]	u_t calc. in $\frac{m}{s}$ [20 °C]	u_t calc. in $\frac{m}{s}$ [750 °C]
Feresint RI 250/2,1	0.0201	0.0148	0.0063	0.80	0.49
Ferelet MT 425/3,0	0.0572	0.0778	0.0332	2.05	1.94
Ferelet RI 425/3,7	0.0600	0.0796	0.033	2.05	1.93
Iron Powder Type 120875	0.0595	0.0794	0.034	1.89	1.88
Tierga ore	0.0530	0.0784	0.0335	2.02	1.94

2.2.2 Oxidation degree

To assess the efficiency of reduction and oxidation, the oxidation degree (OD) is determined based on the mass increase while oxidizing. The oxidation degree indicates the proportion of the bed material sample that is in an oxidized form. For this purpose, the mass of the sample is measured before the oxidation in a muffle furnace. After oxidation in the muffle furnace, the mass can be measured again and the mass difference calculated. Oxidation in the muffle furnace is carried out at 750°C or 900 °C over 24 hours. A sample mass of 5 g is used. To ensure complete oxidation, the sample is oxidized again for 30 minutes in the furnace. After the further oxidation, a counterweighing is performed. The additional mass gain must not exceed 1% of

the mass gain from the first oxidation to be considered fully oxidized. Otherwise, the last step of the procedure is repeated.

The weight increase of the sample, while fully oxidizing, divided by the total possible mass difference, yields the oxidation degree. The chemical composition of the material used is known from the data sheets. For the calculation of the maximum theoretical mass difference, it is assumed that the sample material is completely oxidized after oxidation (iron is present only as Fe_2O_3). With the calculated theoretical difference, the weight of the powder in the fully reduced state can be calculated:

$$m_{\text{reduced}} = \omega_{\text{Fe}} \cdot m_{\text{oxidized}} \cdot \frac{M_{\text{Fe}_2\text{O}_3}}{M_{2 \cdot \text{Fe}}}$$

$$OD = \frac{(m_{\text{sample}} - m_{\text{reduced}})}{(m_{\text{oxidized}} - m_{\text{reduced}})}$$

The reciprocal of the oxidation degree is referred to as the reduction degree (RD), which indicates the proportion of the sample material that is in a reduced form:

$$RD = 1 - OD$$

For more precise determination, three samples are evaluated in each case, and the results are averaged. The data obtained is shown in the Table 1.

2.2.3 Solid density

Solid density describes the true density of a substance; porosity and air trapped within a bulk material are not considered. The determination is carried out in accordance with DIN EN ISO 1183-1 [1] using a liquid pycnometer. Figure 2 shows one of the pycnometers used.

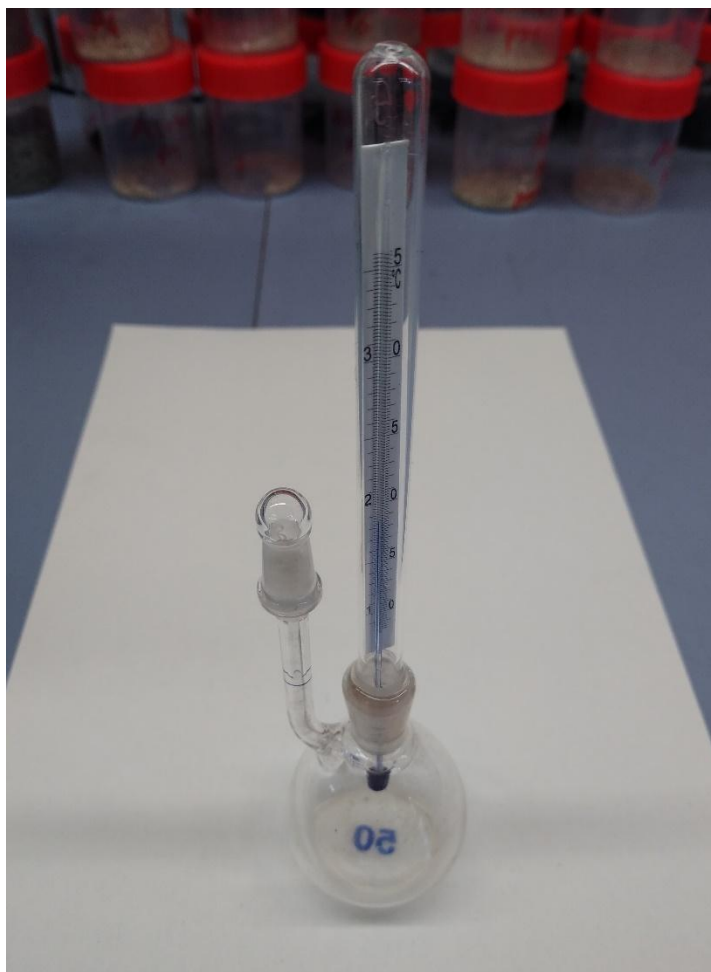


Figure 2 Pycnometer

The device consists of a measuring body, a thermometer and a cap. First, the empty mass of the pycnometer is recorded. About 5 g of the sample are then weighed into the pycnometer and covered with distilled water as the test fluid. The pycnometer is evacuated until no bubbles remain. After degassing, it is filled completely with distilled water, and the thermometer and cap are fitted. The same procedure is repeated without the sample. The density of the sample is calculated with the following equation:

$$\rho_{sample} = \frac{m_{sample} \cdot \rho_{fluid}}{m_{pycnometer, empty} - m_{pycnometer, full}}$$

The density of the immersion fluid (distilled water) is taken from literature values at the measured temperature. For each material, the solid density is determined three times, and the average is used. The data collected is presented in a Table 1.

2.2.4 Particle size distribution

The particle size has a significant influence on the effects examined in this study. As the particle size decreases, the surface-to-mass ratio increases. This improves the reactivity but also increases adhesion forces during fluidisation, making fluidisation more difficult. Therefore, analysing the particle size is an essential basis for the experiments conducted in this work.

Fractionation can be based on various properties, including size (sieving), density (sedimentation), or similar criteria. In this work, the distribution is determined based on mass.



Figure 3 Air Jet used for PSD analysis

Specific air-jet sieves with various mesh sizes were used to separate the particles. Figure 3 shows the setup used. It consists of a sieve that can be swapped out for each fraction. Approximately 30 g of powder is used per sample. Sieving is performed under vacuum, which is generated by a circularly moving suction unit located beneath the sieve.

Mass fractions are determined by measuring the mass difference before and after the sieving process. The sieving proceeds from fine to coarse. Between sieving steps, particles are transferred between the sieves. This process is repeated until no powder remains on the sieve. For evaluation, the mass of powder separated during each sieving step is calculated relative to the total initial sample mass according to the formula:

$$\mu_{fractional} = \frac{(m_{before\ sieve} - m_{after\ sieve})}{m_{total}}$$

The weight fraction is plotted against the mean diameter of the sieve fraction:

$$d_{mean} = \frac{(d_{previous\ sieve} + d_{current\ sieve})}{2}$$

For the cumulative distribution, all smaller fractions are summed up to the current value:

$$V(x) = \sum_{i=1}^x \mu_{fractional}(x)$$

The cumulative curve increases monotonically from 0 to 1. The values determined for the materials examined are shown in Figure 4.

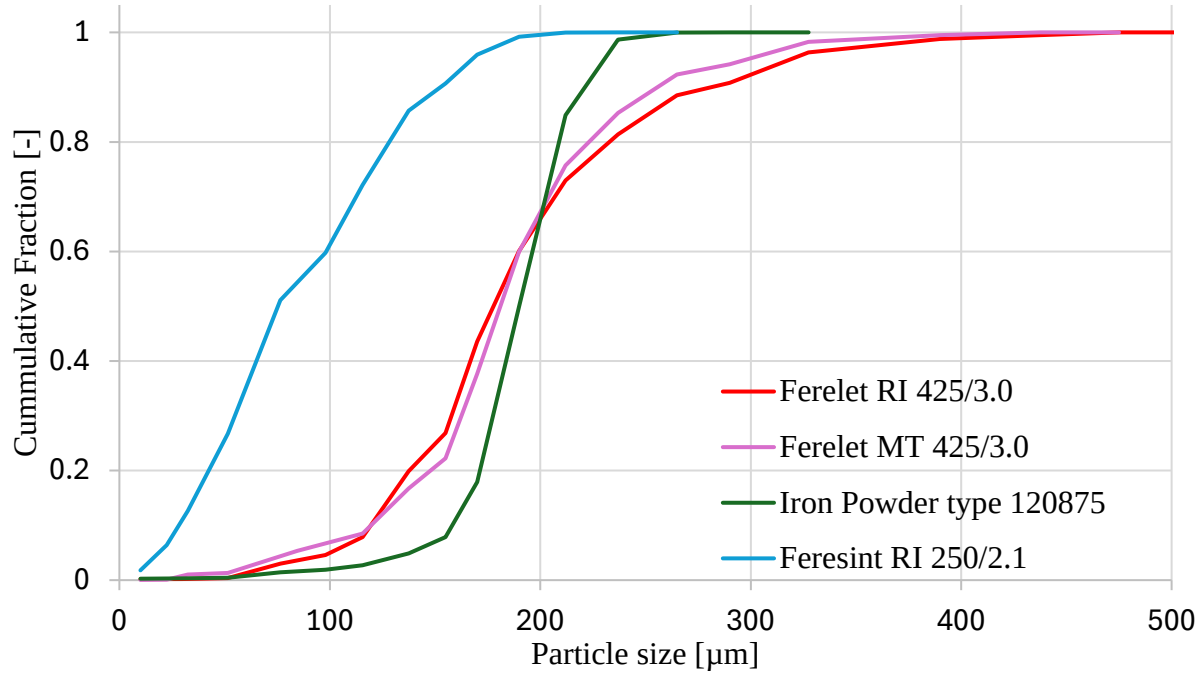


Figure 4 Particle size distribution of the used iron powders

2.3 Fluidisation regime

Due to the design of the test rig, the fluidisation regime investigated in this work is limited to a bubbling fluidised bed (BFB). To ensure this, the fluidisation regime can be illustrated in the Grace diagram. The GRACE graph (named after Levenspiel [3]) is a classification diagram for classifying fluidisation regimes in fluidised beds – depending on particle size and flow velocity. It helps to understand when and how particles are fluidised in a gas stream. The x-axis represents the dimensionless particle diameter d_p^* which incorporates particle size, density, and fluid properties, while the y-axis shows the dimensionless superficial velocity u_0^* relating the gas flow rate to particle and fluid characteristics. By plotting the operating point in the diagram, the corresponding fluidisation regime can be determined, allowing for a direct assessment of the expected bed behaviour under given conditions. Figure 5 shows the fluidisation regime of the materials investigated at an operating temperature of 750 °C and a superficial velocity of 0.5 m/s. For the tests, an attempt was made to select a fluidisation velocity roughly 10 times the experimentally determined minimum fluidisation velocities. For this reason, a superficial velocity of 0.5 m/s was selected and examined for the tested material. For the materials Ferelet MT 425/3.0, Iron Powder Type 120875 and Tierga ore, the operating point is clearly within the BFB range. For the material Ferelet RI 425/3.7 the operating point lies slightly below the BFB range, but above the minimum fluidisation.

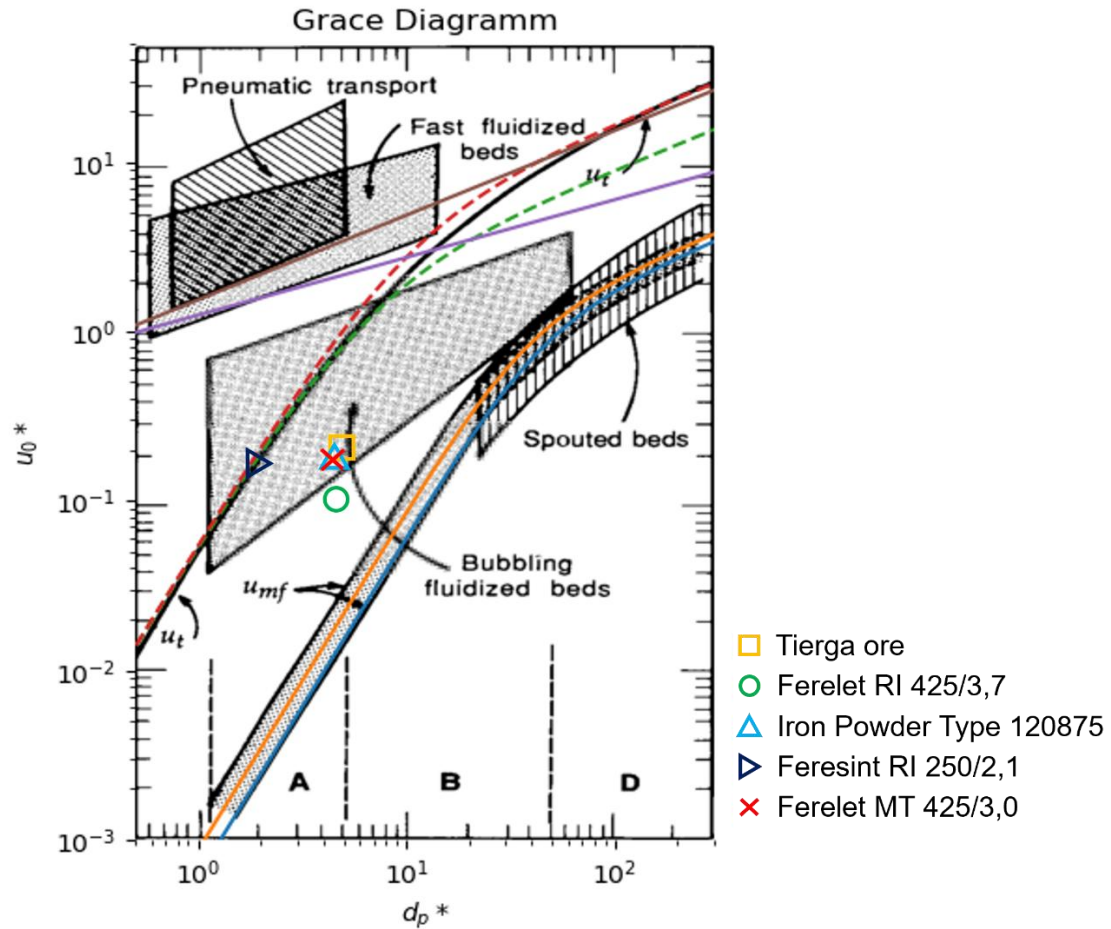


Figure 5 Grace Diagram at 750 °C with 0.5 m/s superficial velocity

When considering the calculated terminal velocity of the iron powder Feresint RI at a test temperature of 750°C, it shows that the fluidisation velocity is greater than the limit value. Therefore, in this case, the fluidisation velocity was limited to a range between 0.1 m/s and 0.3 m/s. Figure 6 shows that for the lower velocity the speed falls below the terminal velocity.

It should be noted that the calculation of the Grace diagram refers to the d_{50} diameter. Finer and coarser particles are not considered. Since the calculations are a mathematical estimate, the value is assumed to be sufficiently accurate.

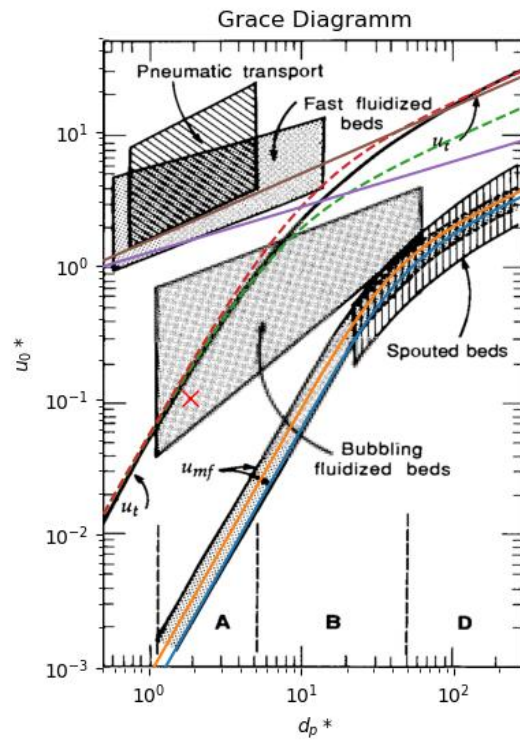


Figure 6 Grace Diagram Feresint RI 250/2.1 at 750 °C and 0,3 m/s

3 Experimental description

This section presents the results used for the planning of the experiments. Particular attention is paid to the properties considered for the material selection and the results on the basis of which the window of operation was decided.

3.1 Setup of the test rig

The experiments were carried out in a bubbling fluidised bed reactor. The structure and possibilities of this setup are described below. Only the properties relevant to the process are discussed. It should be noted that there are several other operating options for the reactor. For operation, basic requirements such as the supply of the operating gases, a power connection, a flue gas duct, etc. must be met. The laboratory fluidised bed is used for the initial assessment of the processes of reactions in a bubbling fluidised bed with low cost and time expenditure. A scheme of the reactor is shown in Figure 7. The components are analysed in more detail in the following sub-sections.

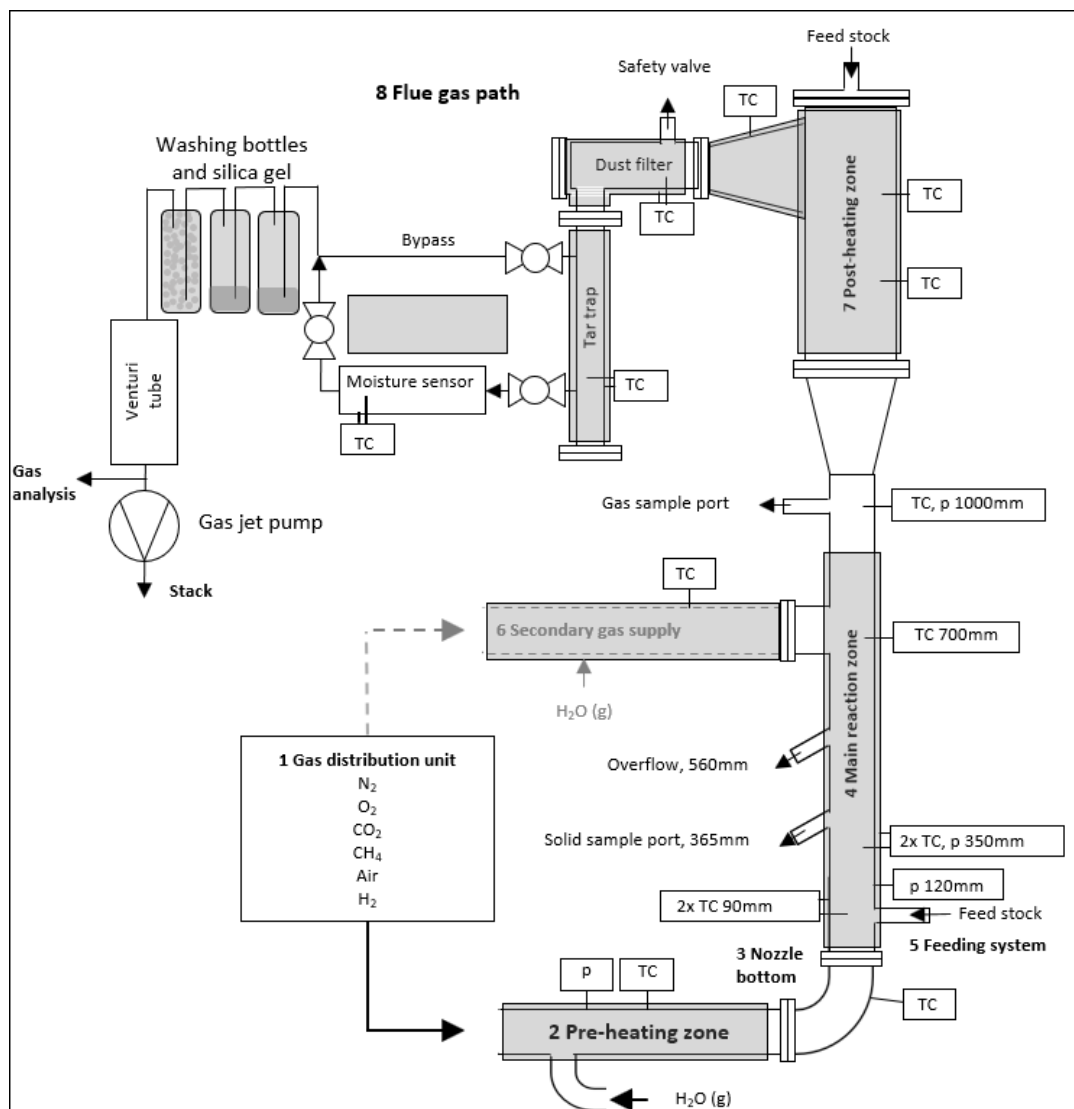


Figure 7 Lab-Scale Reactor Setup

Preheating zone

To operate the fluidised bed at a constant temperature, the gases used in the experiments are preheated before entering the reactor. A preheating zone connects the gas distribution unit with the main reaction zone, enabling control of the temperature of the gases. The temperature of the preheating section can be variably set and controlled via the terminal. The current temperature is measured via a temperature sensor in the preheating unit. The heat supply is generated via an electric heating coil. The preheating section temperature is set to 300 °C.

Main reaction zone

The main reaction zone consists of a continuous reactor tube made of stainless steel (material 1.4841). This has a height of 1000 mm and has a circular cross-section with a diameter of 54.5 mm (DN50). The main reaction takes place in this area. The temperature of the reactor can be controlled via three electrical heating elements. The temperature can be adjusted via a low and high temperature controller. The temperature required for the reaction can be reached and maintained by external heating. In this work, the fluidised bed was only used in batch operation. The complete bed material was already introduced before the reactor was heated up via the ball valve at the top of the reactor. This means that no bed material needs to be added during operation, which simplifies operation. In some cases, additional material can be added through the ball valve on top of the reactor.

Post-heating zone

To reduce the discharge of particles from the reactor, the tube diameter above the main reaction zone is increased to 100 mm (DN100). This leads to a reduction in the gas velocity and thus to less drag forces acting on the solid particles. A large proportion of the particles discharged from the solids bed therefore fall back into the main reaction zone. This effect reduces the share of solids that are entrained in the post-heating zone.

Particle filter

Solid particles that are discharged from the solid bed and do not fall back into the reactor in the reheating zone follow the fluid flow. To separate these particles from the fluid, the flue gas passes through a dust filter. The filter consists of a metal cylinder made of perforated sheet metal fixed in the flue gas path. This is fastened with 4 screws and fitted with a sealing ring to seal the process area from the environment. The cylinder is filled with filter wool. After each test, the filter must be emptied and the wool replaced. This step also removes the entrained particles. The functionality and the amount of discharge in the process can be evaluated after the test by visually assessing the discoloration of the filter wool due to the collected particles.

Distribution plate

As an even distribution of the fluid is advantageous for good fluidisation of the solid material bed, a porous, sintered metal plate with a diameter of 66 mm is clamped at the lower end of the reactor. The plate is positioned between the preheating zone and the main reaction zone. Due to the pressure drop through the plate, the fluid flow is introduced into the reactor in many small, evenly distributed streams. The distribution plate is attached to the reactor with 4 screw connections. Sealing rings are used to seal the process from the environment, which must be replaced each time the reactor is opened. Figure 8 shows the distribution plate used. A hole is provided in the handle of the distribution plate for centring the distribution plate.



Figure 8 Distribution plate of the lab-scale reactor

Solid material extraction

To enable the removal of the solid bed material for further analysis after the experiment, a solids outlet is installed below the reactor bottom. After removing the distribution plate, the bed can flow into the outlet pipe. As an overpressure must be built up below the distribution plate during operation, the outlet pipe is sealed off from the environment by a ball valve. After removing the distribution plate, the reactor inventory can be removed from the reactor by opening this ball valve at.

Gas distribution system

The gas distribution unit is used to feed the gases used to operate the fluidised bed into the process and to control the volume flow. Supply lines are provided for nitrogen (N_2), oxygen (O_2), carbon dioxide (CO_2), methane (CH_4), compressed air and hydrogen (H_2). Mass flow control units (MFC units) are used to regulate the volume flow at.

In addition, a pressure reducer and a solenoid valve are installed in each line for additional interlocking. The MFCs and solenoid valves are controlled via the computer system. As an additional safety measure, manually switchable ball valves are installed for each gas. The gases

are mixed downstream of the solenoid valves. The mixture is then fed to the preheating section. Figure 9 shows a picture of the gas distribution system.

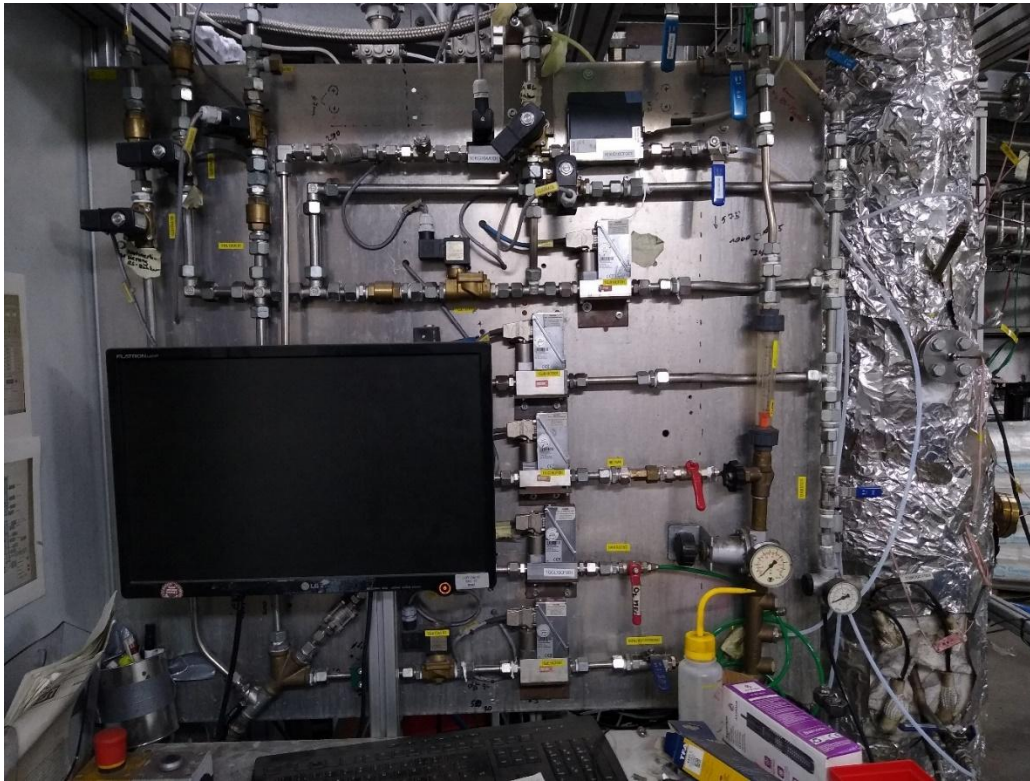


Figure 9 Gas Distribution Unit

3.2 Data recording

Temperature measurement

Since the temperature of the feedstock has an influence on the reaction rate of oxidation and reduction, it is important to know the temperature of the reactor inventory during the test. Therefore, eleven temperature sensors are installed in the test system. Four of the sensors are in the main reaction zone at heights of 130 mm, 350 mm, 550 mm and 1000 mm. The height is measured here in relation to the nozzle base. The other two temperature measurements are taken in the post-heating zone at heights of 1500 mm and 2000 mm. In addition, the temperature is determined in the preheating section of the gases used. The other temperature measurements are taken in the filter, the filter feed, the tar trap (not required in this work), the moisture sensor and the feed of the moisture sensor. These are useful for the operation of the system but are not required for the evaluation of the tests.

Pressure measurement

The presence and operating status of a fluidised bed can be determined via the pressure difference between different heights of the reactor. The necessary differential pressure sensors are installed at three points in the main reaction zone inside and above the solids bed. These are

located at heights of 130 mm, 370 mm and 1000 mm. These sensors can be used to determine the point at which fluidisation takes place.

Sampling of bed materials

Sampling during the ongoing test or between different process steps provides a better understanding of the reactions in the reactor. Two sampling ports are provided on the reactor for this purpose. These are located at heights of 365 mm and 560 mm. In this work, these were only used when switching between reduction and oxidation in the cold reactor state. To remove old particles from the sampling, a flushing valve with nitrogen is attached to the sampling point.

Gas analysis

The fluids converted during the reaction within the reactor are evaluated by calculating the amount of gas remaining in the reactor. The material composition of the exhaust gas is required for the calculation. In this work, the mobile gas analysis unit VARIOluxx from MRU Air is used for this purpose. This can determine the O₂, CO₂, CO and H₂ content of the gas. This is achieved by means of infrared measurement (NDIR) for CO and CO₂ and electrochemical sensor technology (EC) for H₂ and O₂. For this purpose, a flue gas sample is taken before it is discharged from the flue gas outlet. Since nitrogen does not react at the temperatures, it is used to calculate the total volume flow of the flue gas. A special feature of the device is the possibility of measuring hydrogen concentrations up to 100 % by volume. This is of particular importance for the investigations of this work, as it allows a wide range of hydrogen concentrations to be analysed.

3.3 Description of the test campaign

As initial preliminary investigations have shown that iron ore has a lower potential for agglomeration, the first investigations to determine the operating window were carried out using iron ore from the Tierga mine in Spain.

In the experiments, the extent to which the temperature of the experiment influences the degree of reduction achieved was investigated. It was also investigated whether the concentration of hydrogen in the reaction gas has an influence on the reduction. Table 4 and Table 5 show the test matrix of the oxidation and reduction experiments on which this paper is based. In the following chapters the results are only shown exemplary. The process window is only limited by defluidisation of the material or by stopping the reaction.

The experiments were conducted as batch tests in two different ways. On the one hand, pure batch tests and, on the other hand, drop-in batch tests were examined.

In pure batch tests, the reactor is filled with the sample material (1 to 2.5 kg) or a mixture of sample material and inert bed material before the tests. As the experimental preparation the material is heated together with the test rig to the target temperature under inert atmosphere. Afterwards as the experiment starts, the reactor is then charged with the reaction fluid (air or hydrogen). The reaction takes place over a certain period, depending on the and a material sample can be taken while the test is running or after the test is completed.

In the drop-in tests, a small amount of bed material that does not participate in the reaction is added to the reactor and then heated together with the reactor. The sample material is added to the reactor after the target temperature has been reached. After waiting approx. 2 minutes for the materials to mix, the reaction fluid is fed into the reactor. Due to the lower mass of the sample, the reaction takes place more quickly, but it is not possible to take a representative material sample.

The temperature range investigated in this work was between 580 °C and 750 °C. The fluidisation speed was chosen due to the Minimum Fluidisation tests described in 2.2.1. To ensure a stable operation a fluidisation velocity 10x higher than the minimal fluidisation was chosen. Therefore, in the following experimental investigation a fluidisation velocity of $0.5 \frac{m}{s}$ was used.

For the reduction tests the hydrogen concentration was varied in the range between 20 vol.% and 80 vol.%. For the tests with iron ore, the reactor was heated to the target temperature together with the material bed. The fluidisation while heating up consisted of pure nitrogen. At the start of the test, the fluidisation was changed so that the desired hydrogen concentration was present at the entry to the reactor.

For the oxidation tests the oxygen concentration was varied between 5 vol.% and 20,95 vol.%. For the experiments drop in experiments as well as regular batch tests were conducted. The Heating up of the reactor was done similar to the reduction experiments.

During the tests, the conversion of the hydrogen and oxygen were evaluated by means of a continuous gas extraction from the exhaust gas. Nitrogen was used as the tracer gas. The degree of hydrogen conversion can be determined for each moment by conversion. When a conversion rate of less than 10% was reached for batch tests or after a specific reaction time of 20 minutes

for the drop in tests, the experiment was terminated. For the following cooling of the reactor pure nitrogen was used to preserve the oxidation degree of the bed material.

3.4 Occurred limitation of experimental operation

Various limitations were encountered during the experimental investigations in the lab-scale reactor. These are due to both the design of the test rig and the nature of the iron powder.

One of the limitations is the test temperature. The lab scale reactor is electrically heated as described in 3.1. As heat loss occurs via the fluidisation gas and heat losses on the surface of the reactor, there is a limitation to the temperature. During operation with iron powder, this limit occurred at 750 °C. Therefore, the investigations could not be carried out at higher temperatures.

Figure 10 shows the so-called Baur-Glässner diagram [according to [4]]. In this diagram, the test temperature is plotted against the gas oxidation degree (GOD). This indicates the ratio of oxidized fluidisation gas over the sum of oxidized and non-oxidized fluidisation gas. The diagram shows the occurrence of oxidation states of iron as a function of temperature and GOD.

For the investigations in the scope of the CORAL project, the reduction with an intermediate step via Wüstite (FeO) is to be investigated. To ensure this, a temperature must be selected at which Wüstite occurs. According to Figure 10, this is the case from a temperature of approx. 850 K (approx. 580 °C). This temperature thus forms the lower limit of the investigations.

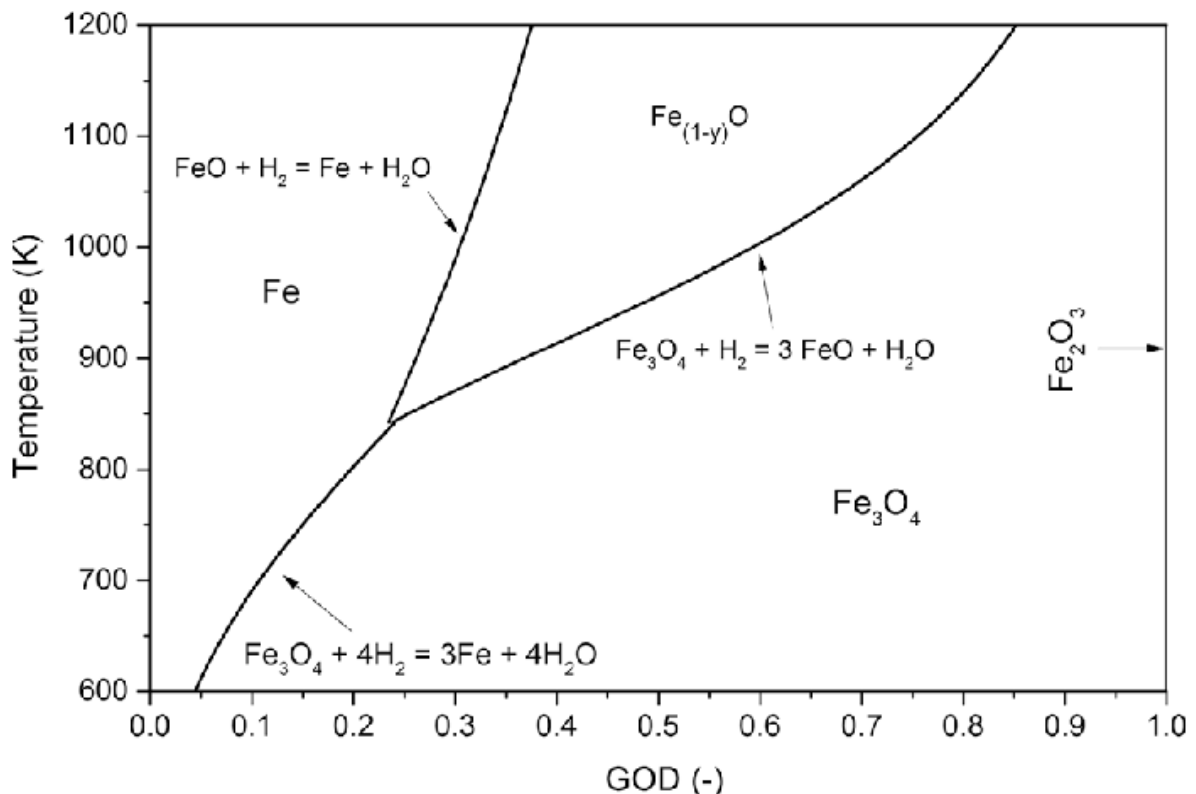


Figure 10 Baur-Glässner-Diagram for Hydrogen Reduction of Iron [4].

A further limitation occurs for the maximum pressure that can be measured. The pressure reducers described in 3.2 have an upper limit of the measuring range at 300 mbar above atmospheric pressure. The highest pressure occurs in the preheating zone. The greatest pressure

losses occur during the flow through the sinter plate of the nozzle base and over the material bed. Based on experience with the lab-scale reactor, the mass of the material bed is limited to approximately 2.5 kg.

A further limitation of the experimental investigations was the agglomeration that occurred in the test and the associated defluidisation of the bed material. The reasons for this could be the high temperature of the experiment or rapid reaction of the material, which leads to very high temperatures at the particles surface. The high temperature might entail a partial melting of the particle and agglomerations due to contact with other iron particles. To reduce this effect, the concentration of the reaction gas was increased in stages or another substance not involved in the reaction (e.g. limestone) was added to the bed material. This additive reduces the number of contacts of reacting iron particles and thus reduces the susceptibility to agglomeration.

In the evaluation of the tests, the limitations were decisive for the choice of the material for the further examination in the project.

4 Results and discussion

4.1 Assessment of iron powders

Several properties were compared to select the iron powder that will be investigated in the further course of the CORAL project. One of the most important requirements here was a broad window of operation and as few complications as possible in conducting the oxidation and reduction tests. Particular attention was paid to the point at which agglomeration occurred in the experiments.

Agglomeration

During the tests, two of the powders proved to be particularly susceptible to agglomeration. This was particularly evident in regular batch tests. Figure 11 and Figure 12 show agglomerations that occurred during the oxidation of Feresint RI 250/2.1 iron powder and type 120875 iron powder. The agglomeration shown in Figure 11 was observed during the oxidation process of Feresint RI 250/2.1, while oxidizing with an oxygen concentration below 5 vol.% in the fluidisation medium at 650 °C. The defluidisation started right after the introduction of the oxygen into the reactor. After examining the bed material, a large, sintered iron piece, as shown in Figure 11, was found in the reactor. The conclusion drawn from this behaviour was that Feresint RI 250/2.1, due to its tendency to agglomerate and defluidise, which occurs even at very low temperatures and oxygen concentrations at the tested scale and therefore was not selected for further testing.



Figure 11 Agglomeration oxidation Feresint RI 250/2,1

Figure 12 shows the agglomeration of iron powder type 120875 inside the tube of the lab-scale reactor. The defluidisation occurred during the heating of the test tube under an inert atmosphere at a temperature of approximately 620 °C. Since sintering occurred even before the reaction began a second test with a mixture of limestone and iron powder type 120875 (20 wt.% limestone) was conducted. The second test showed the same defluidisation at similar temperatures. Due to this behaviour the experiment could not be carried out. For this reason, iron powder type 120875 was also assessed as not suitable for further investigations. During the experiments with the two other iron powders no major agglomerations occurred.



Figure 12 Agglomeration Oxidation Iron Powder type 120875

Reaction evaluation

To further investigate the suitability of the iron powders, drop-in tests were carried out under the same test conditions for the two other iron powders, Ferelet RI 425/3.7 and Ferelet MT 425/3.0. Since the powders are similar products, with the addition of RI for further reduction after production and otherwise similar properties, the reactivity of the two powders is assessed as similar. To verify this, the reacted oxygen is evaluated. Figure 13 shows the course of the absorbed oxygen over time in the test period. The mass during this experiment was 50 g. The curves are plotted on the same starting point with introduction of oxygen in the reactor. Comparing the reacted oxygen is suitable for the comparison because it directly correlates to the change in oxidation degree. The progressions over time are very similar and overlap at multiple points. The two materials seem to have the same oxidation behaviour. Based on these results, the two iron powders appear to be equally suitable.

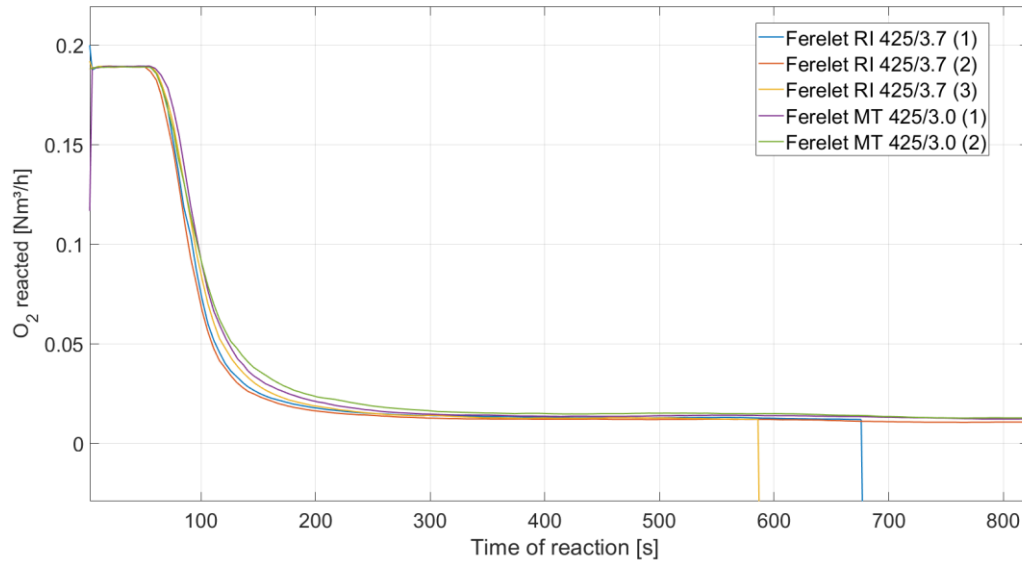


Figure 13 Comparison Ferelet RI 425/3.7 and Ferelet MT 425/3.0

Costs

Another property considered in distinguishing between the two iron powders is the economic efficiency of the process. In this context, low production costs are considered preferable. After consulting with the manufacturer of POMETON iron powder, it was found that Ferelet MT 425/3.0 powder has the lowest production costs.

Table 3 Comparison of the properties of iron powders

Property	Feresint RI 250/2.1	Iron Powder type 120875	Ferelet RI 425/3.7	Ferelet MT 425/3.0
Agglomeration behaviour	- -	-	+	+
Reaction Property			+	+
Production Cost			~	+

The above evaluations of the properties of the iron powders are also shown in Table 3 Comparison of the properties of iron powders. Based on these results, Ferelet MT 425/3.0 powder was selected as the material to be investigated in the CORAL project and was used for the experiments described in 4.2 and 4.3.

4.2 Results of the reduction experiments

Since the reduction of the iron powder could only be carried out after prior oxidation, the results depend on the degree of oxidation achieved. Against this background, experiments were conducted to evaluate the influence of temperature and hydrogen concentration on the reduction of iron ore. The feasibility of reduction under the preferred conditions was then demonstrated using oxidised iron powder.

4.2.1 Iron ore powder

The reduction tests with the iron ore from the Tierga mine were carried out as batch tests. A bed mass of approx. 1-2 kg was placed in the reactor before the system was heated up. During the experiments, the temperature of the reactor and the hydrogen concentration in the fluidisation gas were varied to determine the influence on the reduction.

Figure 14 shows a typical progression of the reduction of iron ore over the test period. The slope of the curve is directly related to the reduction rate. A clear change in slope can be seen at approx. 3700 s. After this point, the gradient is significantly lower. The point at which the gradient changes thus indicates a value from which the reduction requires a much greater effort to further increase the degree of reduction. For this reason, the point is used to evaluate the reduction. The point is given exemplary in Figure 14.

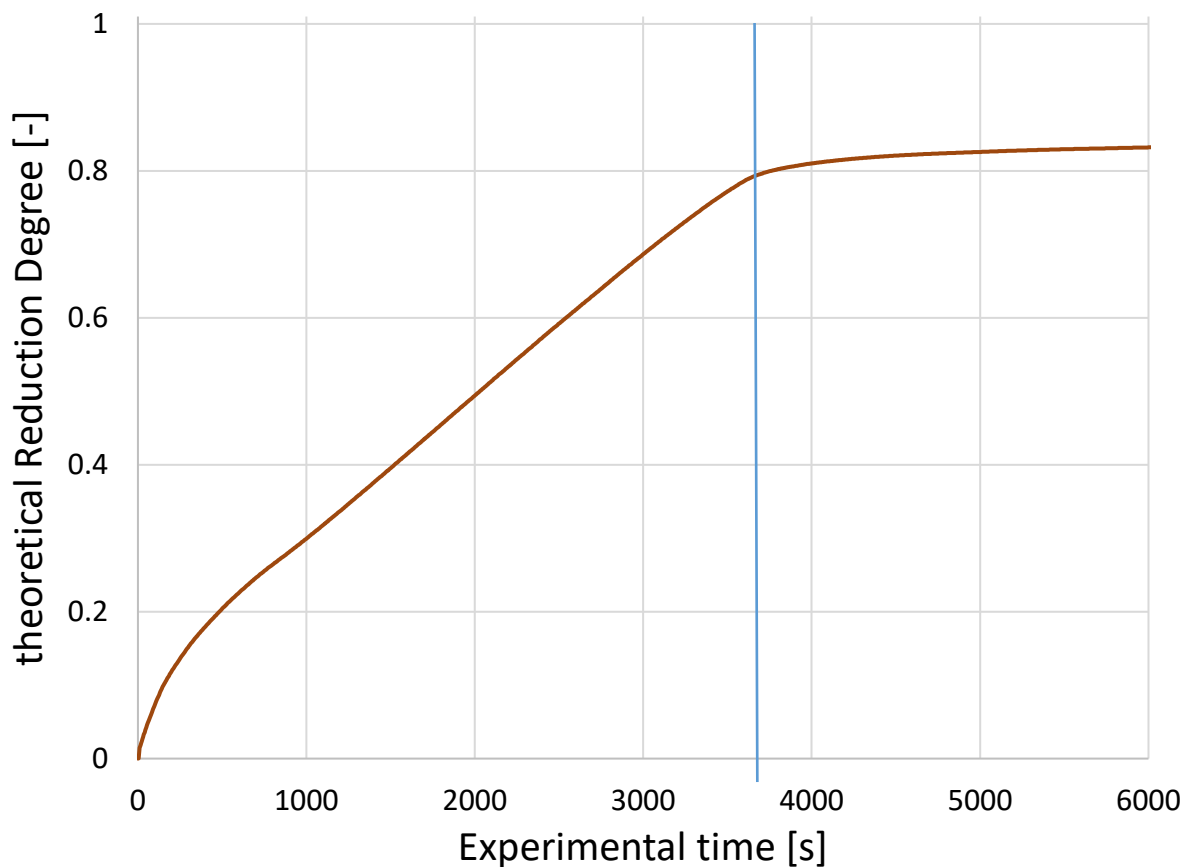


Figure 14 Time course of reduction degree over the experimental time in reduction experiment with Tierga ore at 750 °C and 80 vol.% hydrogen

The dependence of the reduction on the experimental temperature is shown in Figure 16. The temperature varied between 580 °C and 730 °C at a hydrogen concentration of 80 vol.%. The degree of reduction at the point of the change in gradient is used as the evaluation. The test results show a clearly increasing trend. This shows that a high degree of reduction can be achieved with less effort at higher temperatures. The highest possible reactor temperature is then desirable for the reduction.

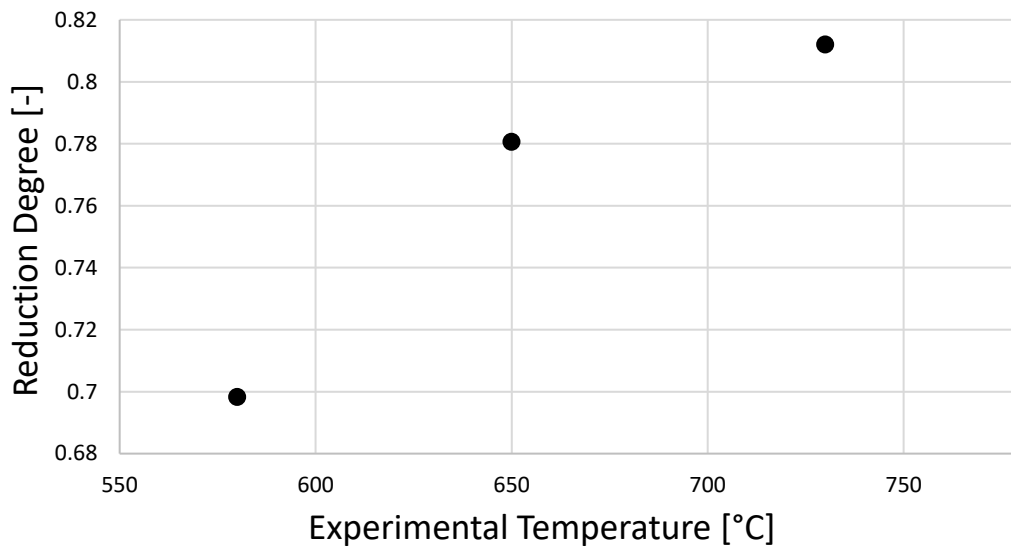


Figure 15 Temperature dependence Reduction at 80 vol.% hydrogen

The hydrogen concentration in the fluidisation gas was varied between 30 vol.% and 80 vol.% using the same procedure to determine the dependence of the degree of reduction achieved at the point of the change in gradient. A temperature of 750 °C was selected based on the previous investigations. The results are shown in Figure 16. The results show an increasing degree of reduction at higher hydrogen concentrations. Compared to the test temperature, however, the influence is smaller. Nevertheless, it is advantageous to select a higher hydrogen concentration.

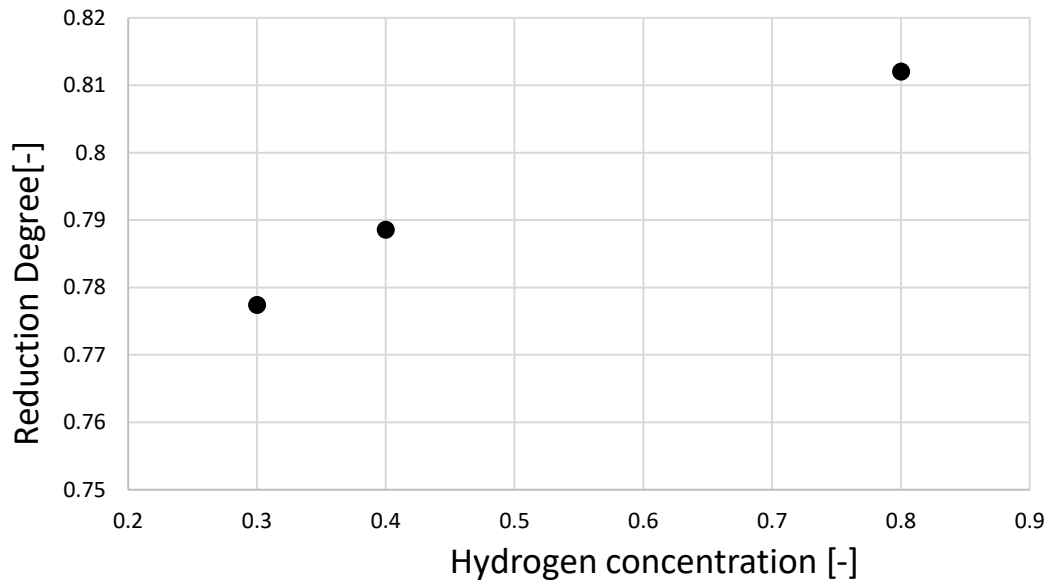


Figure 16 Hydrogen concentration dependence at 750 °C

4.2.2 Iron powder

The iron powder reduction test was carried out for the iron powder selected during the task. The iron powder was selected based on its oxidation behaviour, as described in more detail in the selected powder Ferelet MT 425/3.0 was first oxidised in a regular batch test. The material was then reduced under the conditions determined in the tests with iron ore. The resulting challenges required an adaptation of the test procedure.

The iron powder was mixed with limestone to prevent the particles from agglomerating during the reaction, and the concentration of hydrogen was gradually increased to a maximum value of 32 vol%.

Hydrogen concentration of reduction experiments

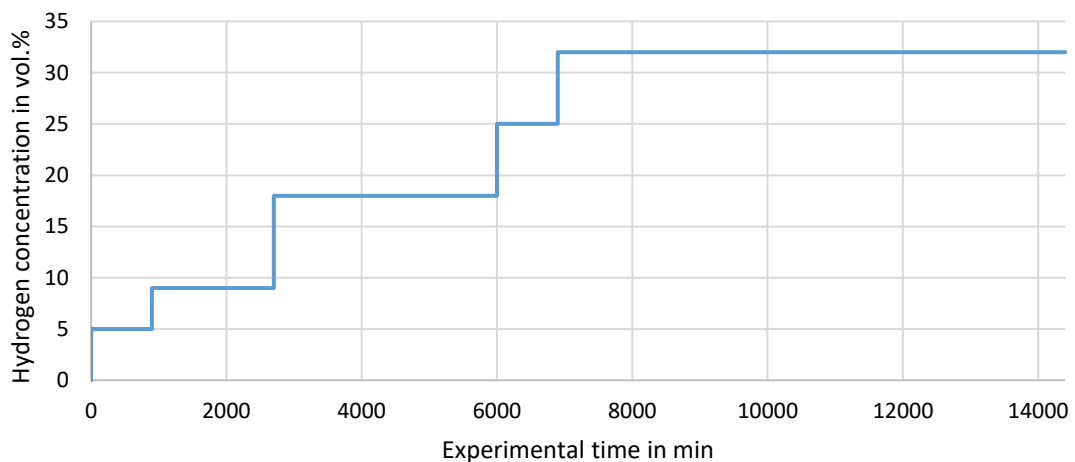


Figure 17 Hydrogen concentration of reduction experiments

Figure 17 shows the gradual increase in the hydrogen content in the fluidisation. The conversion and fluidisation were assessed visually to develop the method. The experiment ended after 240 minutes. During the whole experiment the temperature was kept stable at 750 °C.

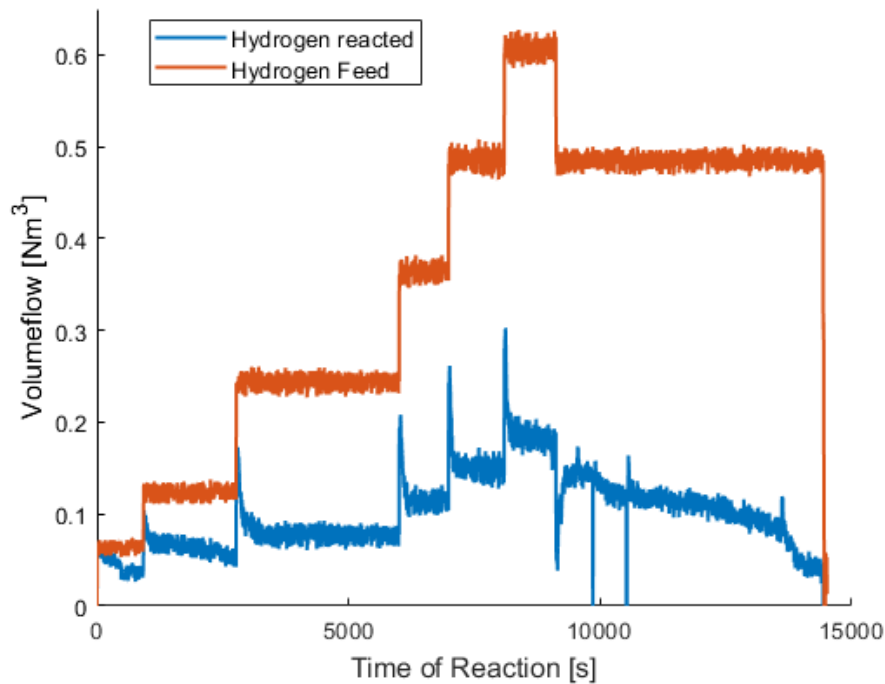


Figure 18 Volume of Hydrogen in feed and reacted

Figure 18 shows the reacted volume flow of hydrogen and the volume of hydrogen fed into the reactor. The feed clearly shows the gradual increase in hydrogen fed to the reactor. The reacted hydrogen volume increases stepwise with the rise of hydrogen concentration in the fluidisation medium. Over the course of the experiment, the reacting volume continues to decrease. For further evaluation, the fraction of hydrogen that reacts is also evaluated in Figure 19. The reacted proportion starts with almost all the hydrogen reacting. In addition, a longer-term phase with uniform conversion can be seen despite an increase in the hydrogen volume flow at approx. 30 vol.%. If this is compared with the estimate from Figure 10, the conversion corresponds to the expected value of GOD for reduction to pure iron at the given temperature. This indicates that the reduction assumptions were correct. From this it can be concluded that a higher temperature as described in the Baur-Glässner diagram leads to a better utilization of the hydrogen. From the results in 4.2.1 it can be concluded that the temperature of the reaction for the reduction is one of the main influences on the efficiency of the reaction.

However, a high temperature also increases the risk of agglomeration, which is why appropriate countermeasures must be taken for the respective material used.

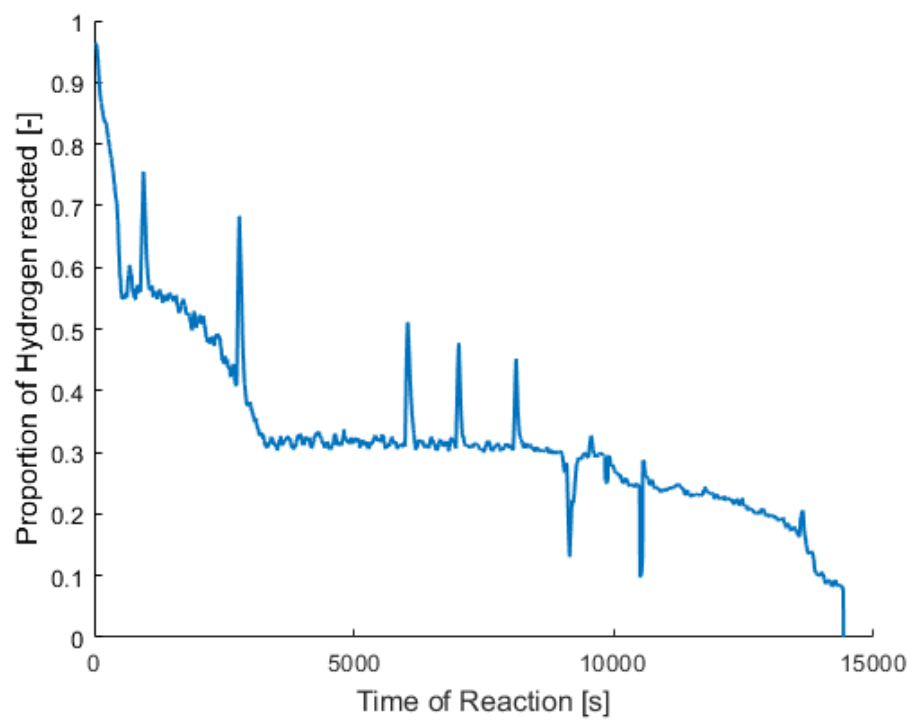


Figure 19 Fraction of hydrogen reacted

4.3 Results of the oxidation experiments

The oxidation tests for the iron powder were carried out as drop-in tests with the selected iron powder Ferelet MT 425/3.0. The temperature of the reactor and the oxygen concentration of the fluidising gas were varied to determine the dependencies of the oxidation process. Compressed air was used to supply the oxygen. As a result, the maximum oxygen content is limited to 20.95% by volume. The maximum temperature is set at 750 °C as the upper limit due to the utilised reactor.

The test temperature is measured over the test period and controlled by the electric heating elements. The results are evaluated using the average temperature over the test period. For the oxidation tests, the influence of the oxygen concentration was examined first. The temperature was kept constant over several tests and the oxygen concentration in the fluidisation medium was varied. Figure 20 shows the temperature curve of the drop-in tests at 700 °C. The heating phase of the reactor and the slight temperature changes caused by the introduction of the sample material and its reaction can be seen. It is also apparent that the temperature at the upper edge of the reaction zone at 1000 mm is already lower. There are no further electric heating elements at this point, which leads to a slight cooling of the reactor. Similarly, the temperature at the distribution plate is lower due to the lower heating power of the preheating section.

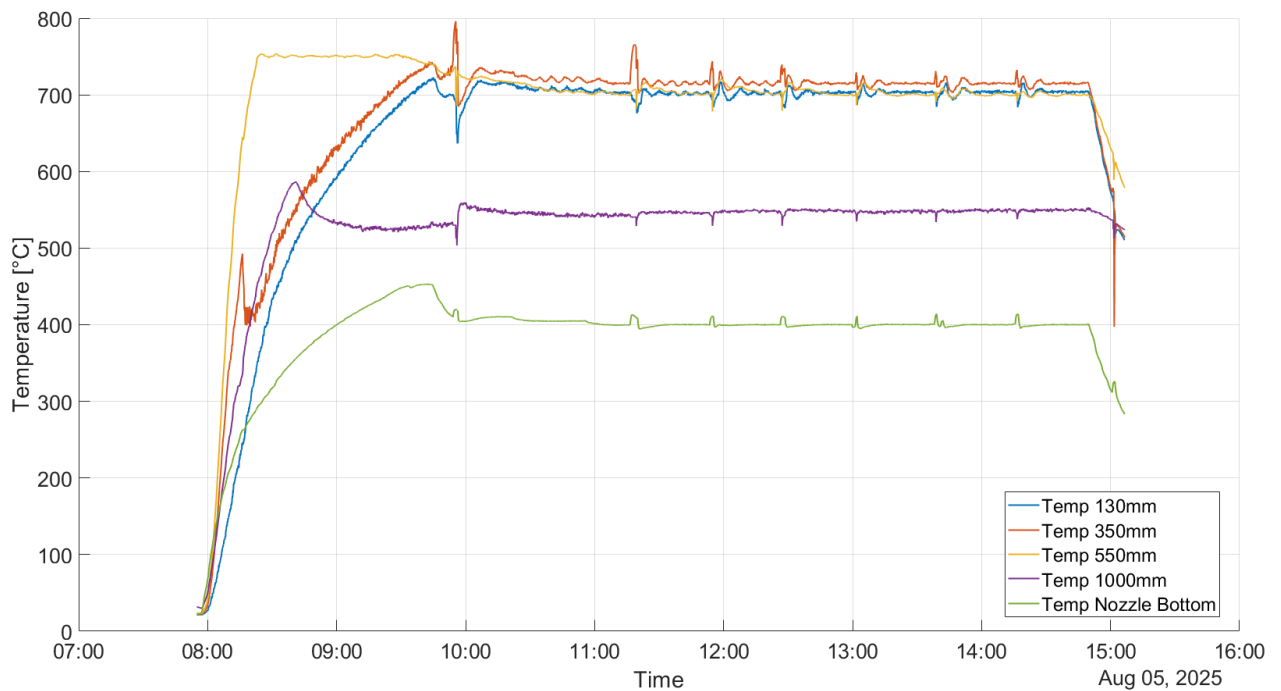


Figure 20 Temperature Drop-In 700 °C

Figure 21 shows the reaction quantities of oxygen corresponding to the temperature curves shown in Figure 20 plotted over the test time. A higher amount of reacted oxygen is equivalent to a faster reaction and higher oxidation degree after a certain time. To achieve reproducibility, the first three experiments from the left were carried out with 100% air and experiments four to six with an air content of 33 % mixed with nitrogen. A higher oxygen uptake can be seen with

a higher oxygen content in the fluidisation medium. It is therefore preferable to use a higher concentration of oxygen in the fluidisation medium for better oxidation and utilisation.

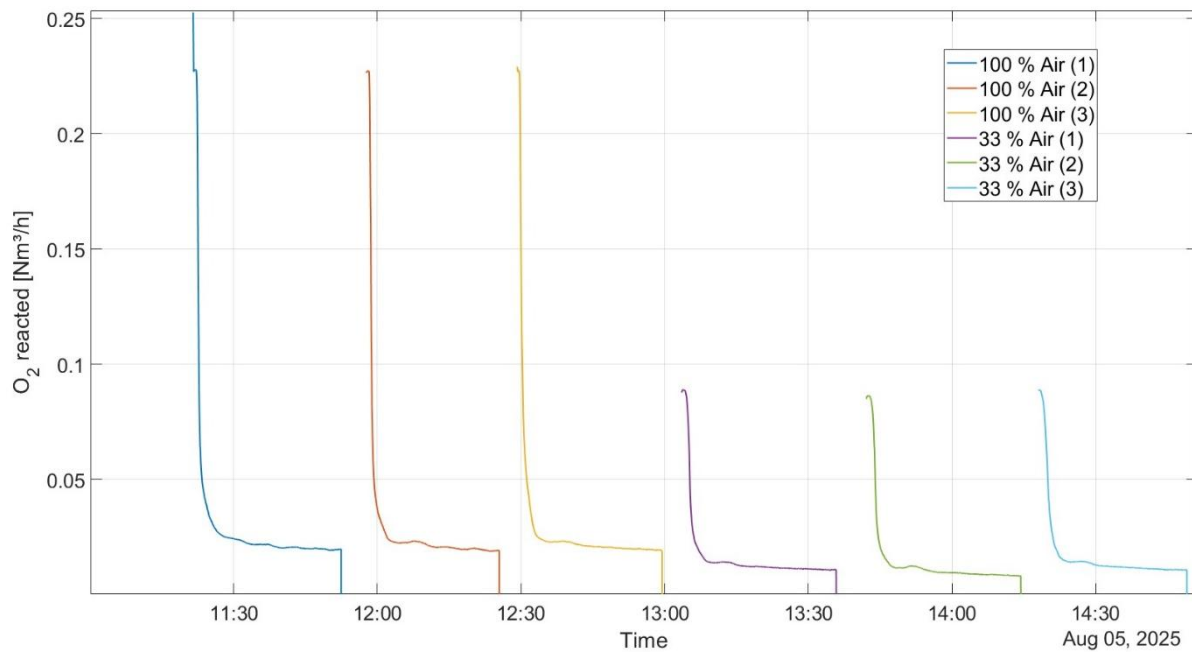


Figure 21 O2 reacted Drop-In 700 °C

A theoretical degree of oxidation can be calculated from the curves shown in Figure 21. The resulting curve is shown in Figure 22. For this purpose, the oxygen consumed is evaluated based on the difference between the inflow into and outflow from the reactor. This estimation leads to a precise understanding of the reaction. The experiment begins with an inert atmosphere in the reactor, which means that the volume of reacted oxygen is overestimated at the start, as there is always a volume of oxygen in the reactor atmosphere. At the end of the experiment, the inflow of oxygen stops, but the gas analysis continues to measure an oxygen content until the previously overestimated portion is compensated for. For this reason, the degree of oxidation drops slightly at the end of the experiment. However, the final value indicates the correct degree of oxidation without overestimation.

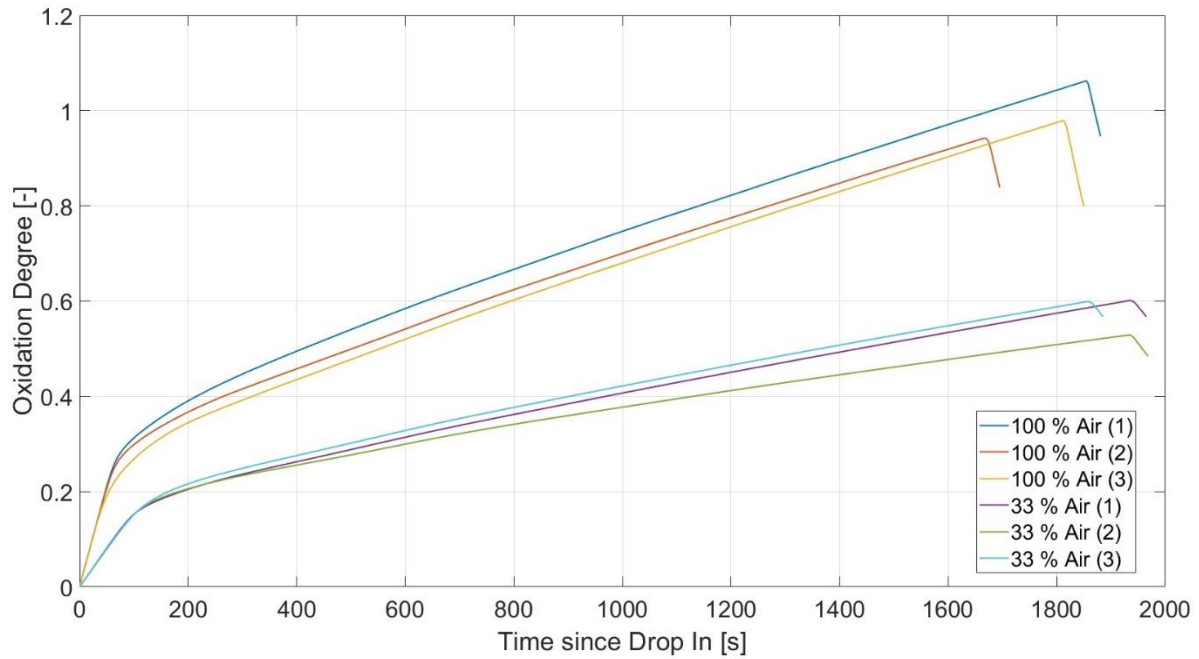


Figure 22 Theoretical Oxidation degree over experimental time at 700 °C

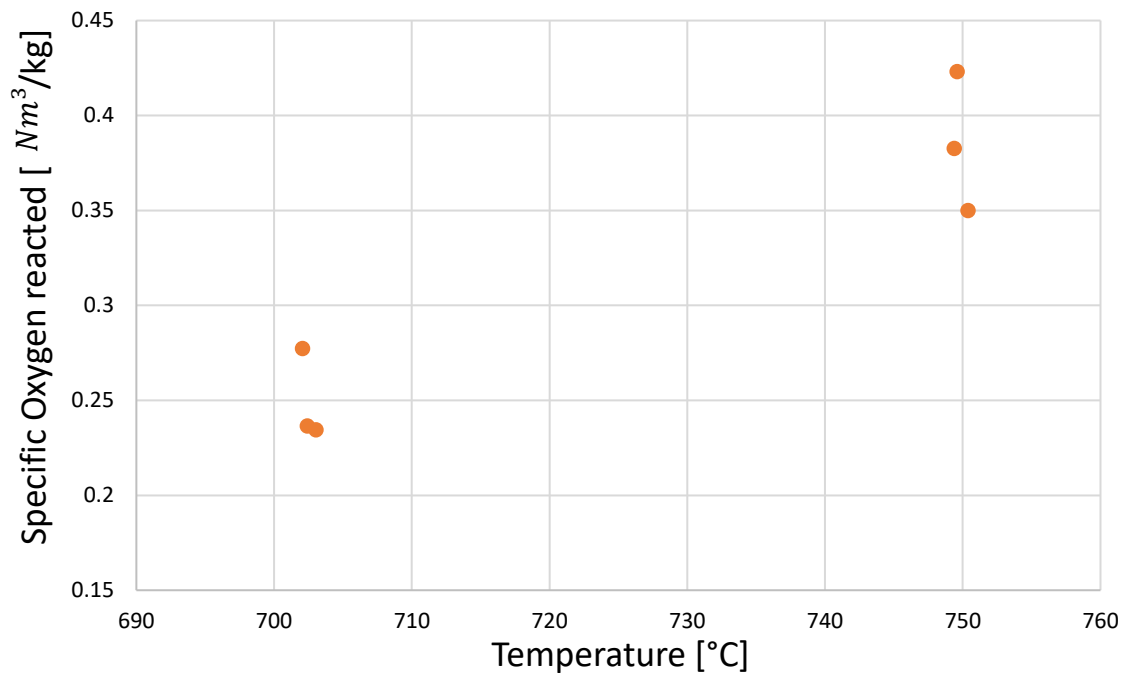


Figure 23 Temperature dependency oxidation with 100 % pressured air

Drop-in tests were also carried out to analyse the dependency of the oxidation reaction in relation to the experimental temperature. The concentration of air in the fluidisation was set at 100 %. Temperatures of 700 °C and 750 °C were selected. The test time was set at 20 minutes for all experiments. Figure 23 shows the results of the tests. The specific amount of oxygen that reacts in the experiment in relation to the sample mass is plotted against the experimental temperature. This representation was chosen because the amount of oxygen reacted is proportional to the increase in the degree of oxidation. A clear trend can be recognised.

According to the results, high temperatures are clearly more suitable for rapid oxidation and are therefore preferable.

5 Conclusion

5.1 Selection of iron powder

The tested iron powders differ strongly in their reaction behaviour and agglomeration tendency. Feresint RI 250/2.1 and Iron Powder Type 120875 show a tendency for sintering, in the latter case already during inert heating. They are therefore unsuitable for further use in the scope of the CORAL project. Ferelet RI 425/3.7 and Ferelet MT 425/3.0 show stable fluidisation and comparable oxidation behaviour without significant agglomeration in batch or drop-in tests. As both are technically suitable, economic aspects guide the choice. Since Ferelet MT 425/3.0 is available at lower cost, it is selected for continued investigations within the CORAL project.

5.2 Determined window of stable operation

Reduction

The stable range extends from about 580 °C, where wüstite appears according to the Baur–Glässner diagram, up to 750 °C, the heating limit of the lab-scale unit. Within this interval, fluidisation remains stable, and agglomeration is avoidable if reactant concentrations are increased gradually and additives such as limestone are applied. Operation near 750 °C accelerates the reaction and enhances conversion efficiency but also raises the risk of sintering, requiring adjusted procedures. Hydrogen concentrations above 30 vol.% clearly improve kinetics and shorten residence times. Although lower concentrations are feasible, the gradual ramp-up of hydrogen during start-up is essential to reduce thermal stress and prevent particle sintering.

Oxidation

Oxidation is limited using air, restricting the maximum oxygen content to 20.95 vol.%. Tests from 600 °C up to 750 °C confirm that higher oxygen levels within this limit lead to faster and more complete oxidation, whereas lower concentrations slow down the process and reduce the overall uptake. In summary, the selected material operates reliably between 600 °C and 750 °C, with flexibility in reactant concentrations. Higher temperatures and concentrations increase reaction rate and efficiency, and with suitable operational measures, these conditions offer significant potential for further optimisation in the CORAL project.

6 References

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7 Appendix

Table 4 Test matrix oxidation

Oxidation			
Material	Air Fraction [%]	Temperature [°C]	Type
Ferelet RI 425/3.7	20-100	650	Batch
Ferelet RI 425/3.7	20-100	725	Batch
Ferelet RI 425/3.7	100	750	Batch
Ferelet RI 425/3.7	100	750	Drop-In
Ferelet RI 425/3.7	100	750	Drop-In
Ferelet MT 425/3.0	100	750	Drop-In
Ferelet MT 425/3.0	100	750	Drop-In
Ferelet MT 425/3.0	100	750	Drop-In
Ferelet MT 425/3.0	5-32	750	Batch
Feresint RI 250/2.1	5-20	600	Batch
Feresint RI 250/2.1	5-10	600	Batch
Feresint RI 250/2.1	5	600	Batch
Ferelet MT 425/3.0	100	650	Drop-In
Ferelet MT 425/3.0	100	650	Drop-In
Ferelet MT 425/3.0	33	650	Drop-In
Ferelet MT 425/3.0	33	650	Drop-In
Ferelet MT 425/3.0	66	650	Drop-In
Ferelet MT 425/3.0	66	650	Drop-In
Ferelet MT 425/3.0	100	700	Drop-In
Ferelet MT 425/3.0	100	700	Drop-In
Ferelet MT 425/3.0	100	700	Drop-In
Ferelet MT 425/3.0	33	700	Drop-In
Ferelet MT 425/3.0	33	700	Drop-In
Ferelet MT 425/3.0	33	700	Drop-In
Ferelet MT 425/3.0	100	750	Drop-In

Ferelet MT 425/3.0	100	750	Drop-In
Ferelet MT 425/3.0	100	750	Drop-In
Ferelet MT 425/3.0	33	750	Drop-In
Ferelet MT 425/3.0	33	750	Drop-In
Ferelet MT 425/3.0	33	750	Drop-In
Ferelet MT 425/3.0	100	750	Drop-In
Ferelet MT 425/3.0	100	750	Drop-In
Ferelet MT 425/3.0	100	750	Drop-In
Ferelet MT 425/3.0	66	750	Drop-In
Ferelet MT 425/3.0	66	750	Drop-In
Ferelet MT 425/3.0	66	750	Drop-In
Ferelet MT 425/3.0	66	700	Drop-In
Ferelet MT 425/3.0	66	700	Drop-In
Ferelet MT 425/3.0	66	700	Drop-In

Table 5 Test matrix reduction

Reduction			
Material	Hydrogen Fraction [%]	Temperature [°C]	Type
Tierga ore	5-20	730	Batch
Tierga ore	80	730	Batch
Tierga ore	0,8	730	Batch
Tierga ore	0,8	680	Batch
Tierga ore	0,4	580	Batch
Tierga ore	0,4	750	Batch
Tierga ore	0,3	650	Batch
Tierga ore	0,8	750	Batch
Ferelet MT 425/3.0	5-32	750	Batch