Green Hydrogen for Low-Carbon Steelmaking

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Summary
The iron and steel industry is responsible for approximately 30 % of the global industrial CO_2 emissions. Achieving 80 % CO_2-reduction by 2050, using the actual production routes without breakthrough technologies, seems to be an unattainable goal. Since there are multiple technological pathways to reduce CO_2-emissions, different steelmaking routes were analyzed in terms of feedstock consumption, energy demand and carbon saving potential.

The direct reduction process may be seen as first step to reduce the CO_2-emissions from primary steel production. The high flexibility of this process allows the gradual substitution of syngas derived from natural gas by hydrogen as a single reducing agent. Model-based calculations for the transformation towards a low carbon iron and steel industry were performed in this research. Therefore, process design models for the natural gas as well as a possible design for the hydrogen based DR-process were developed and evaluated.

The goal of this study was to point out possible CO_2-reduction potentials of the considered routes and the additional energy demand required for the direct reduction with hydrogen. Hence, the availability of sufficient amounts of renewable energy to produce green hydrogen plays a dominant role for the decarbonization of the steel industry.

Key Words
Direct reduction, natural gas, hydrogen, CO_2-emissions, energy demand

Introduction
In order to keep global warming below 2 °C, the European Council reconfirmed the EU objective of reducing greenhouse gas emissions by 80-95 % until 2050 compared to 1990-levels [1]. The iron and steel industry is one of the biggest industrial emitters of CO_2. It is estimated that between 4 and 7 % of the anthropogenic CO_2 emissions are produced from this industrial sector in the EU [2]. About 1.9 tons of CO_2 are generated for every ton of steel produced via the most prevalent steelmaking process, the BF-BOF route [3]. Crude steel production in the EU is almost entirely divided between the BF-BOF and the Scrap-EAF routes, representing respectively 60.5 % and 39.5 % of production shares [4]. The maximum CO_2 reduction potential attainable (considering the decrease of CO_2 intensity of the power sector as well as the increase in scrap availability) with the actual production routes presented in Figure 1 is about 15 % between 2010-2050 [3].
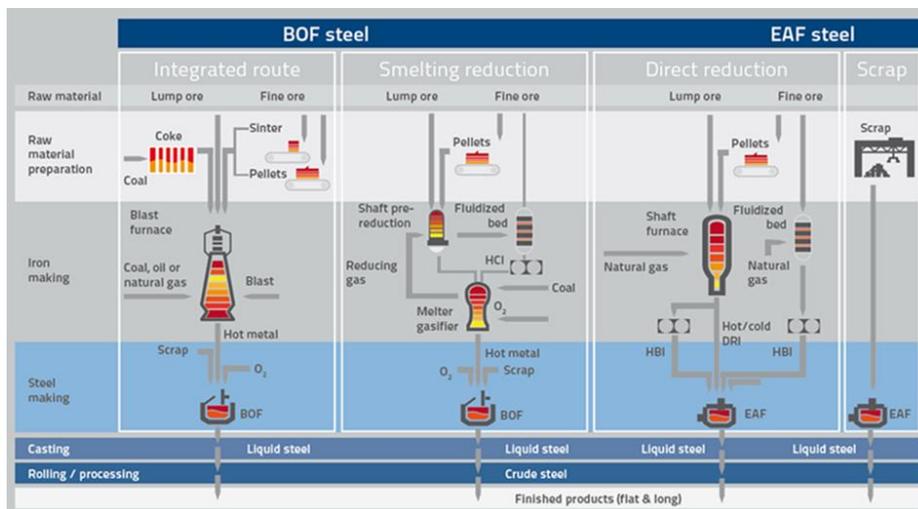
Figure 1: Overview of iron and steel making routes [3]

Because of its low carbon footprint the scrap-EAF route is clearly in advantage, however natural iron sources as raw material will still be required in future due to the limited availability of scrap. This makes the 80 % CO_2-abatement goal unachievable without the use of the so-called "breakthrough technologies".

As depicted in Figure 2, there are different technological pathways to substantially reduce CO_2 emissions in the steel sector: (i) Carbon Capture and Usage (CCU), CO_2 captured is used as a raw material for chemical conversion, (ii) reduced use of carbon in combination with Carbon Capture and Storage (CCS) and (iii) Carbon Direct Avoidance (CDA), directly avoiding CO_2 emissions through an increased use of renewable electrical power in steelmaking to replace carbon [5].

	Smart Carbon Usage (SCU)		Carbon Direct Avoidance (CDA)	
Pathways	Process integration with reduced use of carbon (+CCS)	Carbon Valorisation/Carbon Capture and Usage (CCU) (+CCS)	Hydrogen	Electricity
Description	Process integration with reduced use of carbon	Using CO/CO$_2$ from steel mill as raw material (Chemical Conversion of CO/CO$_2$)	Use of renewable electricity in basic steelmaking, e.g. production of H$_2$ to replace carbon	

Figure 2: Technology pathways for CO_2-reduction [5]

Electrical power-based iron reduction technologies have gained increased attention due to its high CO_2 reduction potential. Up to 95 % of CO_2 can be avoided by using 100 % external green electricity either for direct electrolysis processes, currently being in state of development, or for the production of hydrogen and replacement of carbon-containing reducing agents [2].

As seen in Figure 1, the direct reduction with natural gas (DR-NG) is one of the state-of-the-art steelmaking technologies. DR-NG already operates with approximately 55 % of hydrogen in the reducing gas, coming from the natural gas. Therefore, this process has the potential to gradually introduce additional hydrogen to achieve a further CO_2-reduction. However, there is little information about the behavior of the process when fully operated with hydrogen. Thus, the scope of this paper is to present a potential design of the hydrogen based direct reduction process (DR-H$_2$) and evaluate the energy use and CO_2-emission mitigation potentials.

Natural gas and hydrogen based direct reduction process

Producing steel via direct reduction in combination with an electric arc furnace (EAF) is one possibility for carbon-lean steelmaking. A reduction of about one third of the CO_2-emissions depending on the CO_2 intensity of the electricity mix can be achieved compared with the BF-BOF route [6]. The higher content of hydrogen in natural gas in comparison with coal based reducing agents is directly linked with the decrease in carbon emissions.

There are two dominanting natural gas based direct reduction processes: HYL/Energiron and MIDREX, representing the latter one approximately 65 % of the total worldwide DRI produced [7]. Due to its high implementation grade, a MIDREX process has been selected as the initial point for the calculations.

In the DR process, the iron ore is reduced to solid sponge iron in a shaft furnace by using natural gas as source for the reducing agent. To generate the gases required for the reduction (mainly CO and H$_2$), the natural gas must previously be reformed to syngas. A steam methane and dry reforming process will be simultaneously carried out in the reformer at 900-950 °C, producing a syngas containing about 55 % of hydrogen and 35 % of carbon monoxide according to the following reactions [8]:

$$CH_4 + CO_2 \rightarrow 2\ CO + 2\ H_2 \qquad \Delta H_R = 247\ kJ/mol \quad (1)$$
$$CH_4 + H_2O \rightarrow CO + 3\ H_2 \qquad \Delta H_R = 206\ kJ/mol \quad (2)$$
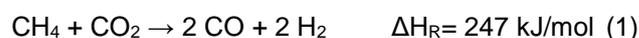
The main chemical reactions for the reduction (removal of oxygen from iron ore) and the carburization (addition of carbon to iron) in the shaft furnace take place as follows:

Reduction:
$$Fe_2O_3 + 3\ H_2 \rightarrow 2\ Fe + 3\ H_2O \quad \Delta H_R = 99\ kJ/mol \quad (3)$$
$$Fe_2O_3 + 3\ CO \rightarrow 2Fe + 3\ CO_2 \quad \Delta H_R = -24\ kJ/mol \quad (4)$$

Carburization:
$$3\ Fe + CO + H_2 \rightarrow Fe_3C + H_2O \qquad\qquad (5)$$
$$3\ Fe + CH_4 \rightarrow Fe_3C + 2\ H_2 \qquad\qquad (6)$$
$$3\ Fe + 2\ CO \rightarrow Fe_3C + CO_2 \qquad\qquad (7)$$
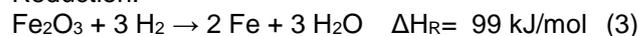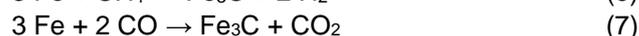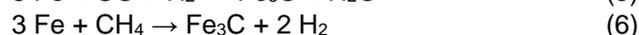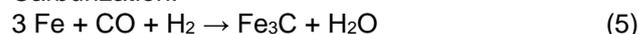
The oxygen from the iron ore reacts with CO and H$_2$ at elevated temperatures to produce metallic iron while releasing CO_2 and H_2O.

An appropriate H$_2$/CO ratio in the reducing gas allows a thermally balanced reduction because the reaction of iron oxides with CO (eq. 4) is exothermic, whereas the reduction with H$_2$ (eq. 3) is an endothermic reaction. The heat required for the reduction with hydrogen is supplied by the heat from the reaction with carbon monoxide.

The remaining off-gas from the shaft furnace, the so-called "top gas" rich in CO_2 and H_2O is treated in a gas scrubber, where water is partly condensed and the dust is removed. Two thirds of those processed gases return as feed inlet to the reformer, once blended with fresh natural gas. The remaining part is mixed as well with natural gas and combusted with air to serve as heating source for the reformer [9].

Hydrogen can be used to partly replace natural gas in the direct reduction process as the reforming gases already account with 55 % of hydrogen and thus

achieve a further reduction in CO_2-emissions. Therefore, no process changes for existing plants are required, up to a substitution rate of about one third of the required natural gas [10].

When using H_2 as reducing agent, a higher reduction degree from iron ore to iron is achieved. Nevertheless, the reduction process is thermally unfavorable, due to the endothermic nature of the reaction between hydrogen and iron oxide [11].

Additionally hydrogen can be used as single reducing agent in the direct reduction process. The top gas, this time mainly composed of H_2O, is partly condensed in the scrubber and removed from the system. Since in this case, no reformer is required, a reduction gas heater is attached to the process to heat the gas to the required temperatures. This heater can be fueled either by hydrogen, natural gas, electricity or waste heat [10].

The temperature in the shaft furncace decreases driven by the endothermic nature of the reaction between iron oxides and hydrogen (see eq. 3). This situation can be dealt with by addition of CH_4 to the process. According to the carburization reactions (eq. 5-7), the natural gas provides carbon to maintain the desired carbon content in the direct reduced iron (DRI). As most of the DRI is used in EAF`s, a certain carbon content must be assured for the formation of a foamy slag and to provide energy to the EAF [8]. Burning the carbon by injecting oxygen in the EAF reduces the electricity consumption and thus enables a faster melting of the charged materials [10].

The prefered amount of carbon in DRI is about 1.5-3 %. Nevertheless, it deviates significantly with the material mix input and the produced steel grades. [10] reports that the addition of 50 m^3_{STP}/t DRI of natural gas leads to a DRI carbon content of about 1.4 % and the temperature levels in the shaft furnace will be mantained.

Currently there is limited information available in literature about the hydrogen demand for a fully hydrogen operated direct reduction process. Due to distinctions in process concepts (e.g. configuration for heaters) there are different values indicated.

MIDREX reports a total hydrogen demand of 800 m^3_{STP}/t DRI. Wheras 550 m^3_{STP}/t DRI are needed for the reduction process itself, 250 m^3_{STP}/t DRI of hydrogen are required as fuel for the reduction gas heater. This amount could also be replaced by alternative energy sources like waste heat, electrical energy and/or natural gas. As mentioned above 50 m^3_{STP}/t DRI of natural gas are used additionally to maintain the temperature and carbon content [12].

Vogl et al. [13] developed a model based calculation for a hydrogen based DR-process in combination with an EAF. They reported a hydrogen demand of 51 kg/t of steel output which is equivalent to about 520 m^3_{STP}/t HBI. According to their description, no additional heat source is required because the reducing gas is heated up by the recoverd heat from the condenser. Additional energy is considered for pre-heating the iron ore.

ArcelorMittal Hamburg GmbH runs the only DR-plant in Europe with a production of approx. 0.55 Mt of cold DRI per year. Based on their process data and experience they also carried out simulations [14]. An overall hydrogen demand of 635 m^3_{STP}/t HBI was reported, including energy losses. The breakdown of the energy requirements is the following: 3 MWh/t HBI for the hydrogen generation and aditionally 0.23 MWh_{el}/t HBI for the electrical heating of the reducing gas.

Modelling approach

In order to assess the hydrogen based direct reduction technology, process models were developed to compare natural gas based process concepts with the implementation of hydrogen. As voestalpine is operating a MIDREX DR-plant in Texas, the layout of the developed process is based on configurations given in [10] and [12]. The aim of the calculations is to identify carbon flows in the DR-process as well as to point out CO_2-reduction potentials and energy demands. The models were set up in a process simulation platform using the m.SIMTOP® model library for metallurgical processes which has been developed by Primetals Technologies and voestalpine in recent years [15]. The system boundaries are chosen as depicted in Figure 3 and Figure 4.

The process models include several material sources and sinks as well as gas splitters and mixers. In addition to this, they include the main unit operations of the processes such as DR shaft, top gas scrubber, product gas compressors and heat exchangers. The reformer model of the (natural gas based) DR-NG process is exchanged by a combination of a gas burner, mixer and heat exchanger model representing the reduction gas heater of the (hydrogen based) DR-H_2 process.

Iron carriers like pellets and lump ore are introduced into the processes by ore source models. Additives like lime are supplied by a separate additive source model. Gaseous inputs such as natural gas, hydrogen or seal gas are implemented through gas source models. The main outputs of the shaft furnace model are hot direct reduced iron of approx. 700 °C and top gas. In order to get comparable results, the same solid material compositions and amounts were chosen for both process models. The shaft furnace itself is simulated as a two zone model with heating and reduction zones which include the reactions given in eq. (3)-(7).

Figure 3: Process model for (natural gas based) DR-NG process with addition of hydrogen

Figure 4: Process model for (hydrogen based) DR-H$_2$ process

The assumed reduction degree for both process models is 96 %. A target value of 1.7 % for the carbon content of the produced DRI is set.

Natural gas can be injected at different process stages: NG as input to the reforming gas, as energy input for heating the reformer or reduction gas heater as well as directly to the bustle gas before entering the shaft furnace. An input of hydrogen in the DR-NG process model is foreseen in the reforming gas before the reformer. In the DR-H$_2$ process model, hydrogen is added to the reduction gas and to the fuel gas for heating of the reduction gas heater. The energy supplied to the heater can also be provided by natural gas.

Results and discussion

In the following section, the obtained results and an evaluation thereof in terms of energy demand, CO_2 savings etc. are presented. As alredy mentioned previously, two different process models were set up: One natural gas based model with the possibilitiy for a gradual introduction of hydrogen into the system and a second one for a hydrogen based DR-process. In order to evaluate the utilization of hydrogen as reducing agent the DR-NG process working exclusively with natural gas was considered as reference case.

1.4 tons of iron ore/pellets are needed to produce one ton of DRI. This amount of iron containing materials is the same for the DR-NG as well as the DR-H_2 process. The calculated CH_4-demand for the DR-NG reference process is 255 m³$_{STP}$/t DRI. In the model it was assumed that natural gas contains approximately 95 % methane (CH_4). The given value is in line with available data from literature which is about 262 m³$_{STP}$/t DRI [16].

Different steps for the addition of hydrogen are calculated based on the DR-NG model. The first step was to replace 30 % of the natural gas by hydrogen. This results in a NG demand of 178 m³$_{STP}$/t DRI and a hydrogen demand of 260 m³$_{STP}$/t DRI (see Figure 5). Theoretically also more hydrogen can be added to the DR-NG process without any process changes. The simulated model compiled up to a $H_2/(H_2+CH_4)$ volume ratio up to 0.87.
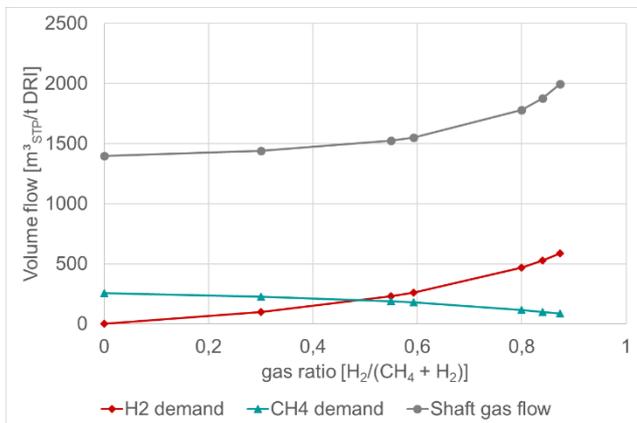
Figure 5: Reduction gas demand for DR-NG process including hydrogen-addition

The substitution of methane by hydrogen is directly connected with an increase of the amount of gas in the system. Approximately four times more volume of hydrogen are necessary to obtain the same metallization degree (amount of oxygen removed from the iron oxide), when substituting methane for hydrogen. As it can be seen in Figure 5 the shaft gas flow exceeds 1,500 m³$_{STP}$/t DRI at a hydrogen share of about 40 %. Therefore, higher velocities and different process conditions in the shaft furnace have to be considered. Despite this, also other process steps

could be affected by varying gas compositions and amounts:

(i) Higher pressure drop inside the shaft furnace and an increase of the gas velocity. These two factors could lead to modifications on the operating conditions, which could affect the behaviour of the reactions inside the furnace.

(ii) Variations on the temperature, composition of the gas and metallization degree because of malfunction of the scrubber. Due to the increase of the gas flow, it may happen that the top gas is not able to cool down sufficiently in the scrubber. Thus, the temperature will be higher and the amount of water in the processed gases will increase, because the water vapour was not able to condense. As the share of water vapour rises, the gas composition will be affected. Therefore the metallization degree of the produced DRI could also be influenced, because these variations can lead to a different potential of the gas to reduce iron oxide.

In principle, the hydrogen can be added at different process stages. In the system configuration defined by MIDREX, hydrogen is pre-heated in a spare unit and directly added to the bustle gas, right before entering the shaft furnace [10]. For the presented calculations hydrogen was added to the reducing gas before entering the reformer. Therfore, no separate heating unit for hydrogen is required. However, the kinetics of the reforming reactions (eq. 1, 2) could be slowed down as a consequence of the increase of the hydrogen volume in the reformer pipes.

By the increase of the hydrogen amount in the shaft furnace, the equilibrium of water gas shift reaction (eq. 8) is also affected due to the principle of Le Chatelier. As more hydrogen will enter to the system, the equilibrium will move to left, generating more CO which will improve the carburization reactions (eq. 5-7).

$$CO + H_2O \rightarrow CO_2 + H_2 \qquad (8)$$

The second model (DR-H_2) was calculated for an input of about 95 % hydrogen. A small amount of natural gas is still considered to maintain process temperatures and the carbon content of the DRI. Other process conditions e.g. amount and composition of input materials, DRI-temperature etc. were not modified. For this process configuration 724-767 m³$_{STP}$/t DRI of hydrogen, depending on the assumed content of hydrogen in the bustle gas, and additionally 48 m³$_{STP}$/t DRI of natural gas are required which slightly deviates from the data given in [10]. Reasons for this could be, that in the presented model different boundary conditions are applied for the simulation and no losses e.g. seal gas losses etc. are considered which will require higher amounts of reducing gas and hydrogen accordingly.

For the reduction of iron oxides itself the demand of hydrogen is 482-525 m³$_{STP}$/t DRI. The remaining part

is needed for heating up the reduction gas and to compensate temperature losses due to the endothermic hydrogen reduction reaction (eq. 3). As stated in [10] it is also possible to provide this amount of energy by alternative energy sources.

Considering the fact that the hydrogen used for the DR-process has to be generated from renewable sources to obtain a carbon-lean steel production, the electrolysis of hydrogen currently seems to be the only way to produce green hydrogen. With an efficiency of 75 % based on the higher heating value of hydrogen (3.54 kWh/m³$_{STP}$) an additional eletric energy demand

between 3,400 and 3,600 kWh/t DRI is required for the production of hydrogen used in the DR-H$_2$ process, including the generation and the heating of the reforming gases. However, in order to increase the efficiency of the process, this heating can be accomplished as well by waste heat from other processes. In this case, the electricity needs, which are limited to the production of hydrogen, decrease until 2,300 to 2,500 kWh/t DRI. This represents the value required for the reduction process itself. The electricity demand for auxiliaries like water and air system etc. is about 80-125 kWh/t DRI [6], [14].
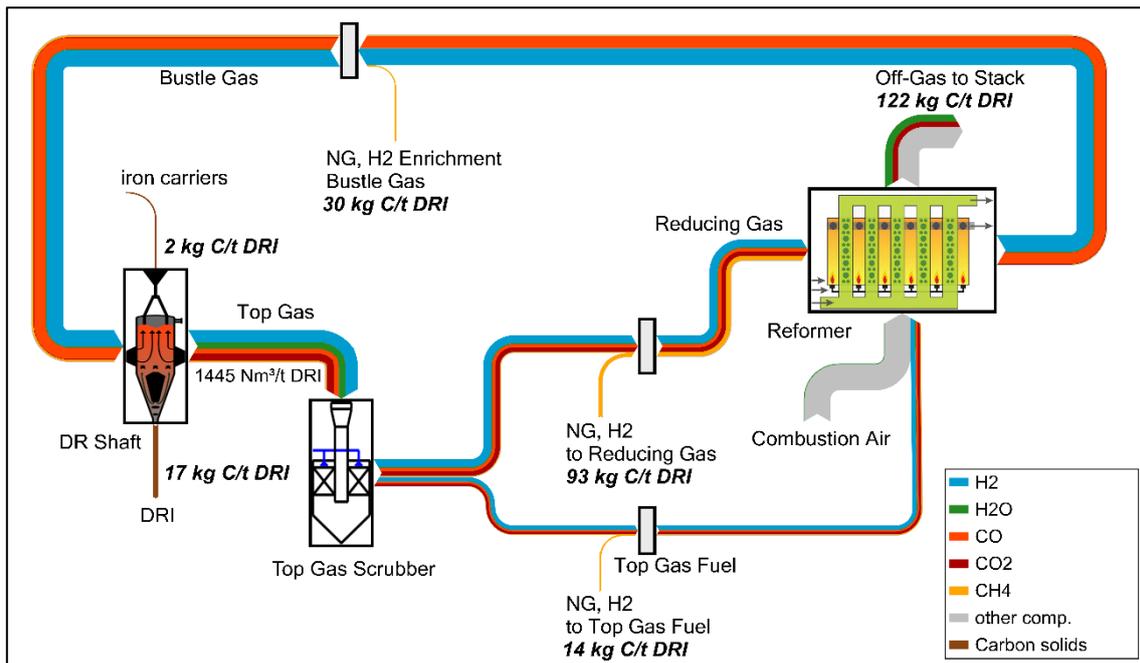
Figure 6: Gas [m³$_{STP}$/t DRI] and carbon [kg C/t DRI] flows for DR-NG process
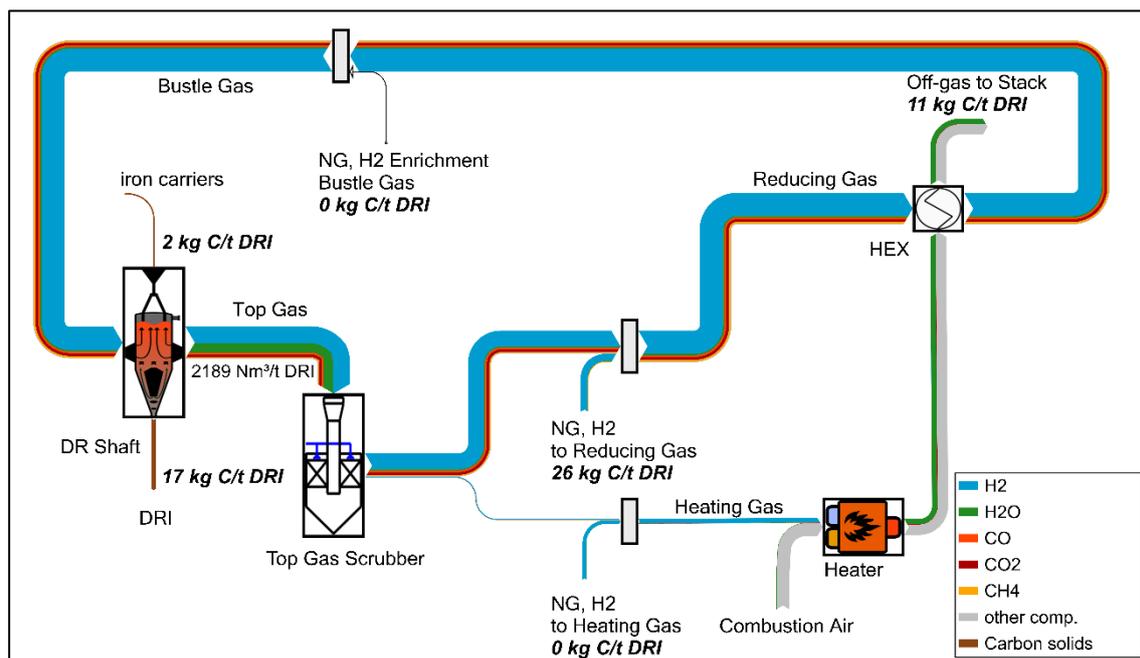
Figure 7: Gas [m³$_{STP}$/t DRI] and carbon [kg C/t DRI] flows for DR-H$_2$ process

The direct reduction process based on hydrogen is one of the future steel production routes with the highest potential for a substantial reduction of CO_2 emissions. In order to evaluate this potential the flows of the different gas components as well as the carbon input and output streams of both process models (DR-NG and DR-H_2) are quantified.

Figure 6 and Figure 7 represents the transition from the DR-NG process with a CO-/H_2 based reduction gas (bustle gas) to the DR-H_2 process where carbon containing gas streams only play a secondary role. According to the process set-up a higher recirculation rate is required for the DR-H_2 model to maintain the desired process temperatures. As a consequence residual amounts of CO and CO_2 are still present in the recycled gas while the overall specific volume flow increases.

As illustrated the main carbon input in the DR-NG process is via the natural gas used as reducing gas or heat input. Here, about 445 kg CO_2/t DRI which are equivalent to about 122 kg C/t DRI are emitted as off-gas from the Reformer. For the DR-H_2 case the principal carbon input is via the natural gas required for the carburization of the DRI. The remaining carbon is released by using the top gas as combustion gas for the heater, representing 39 kg CO_2/t DRI. Thus, up to 91 % of direct CO_2-emissions of the direct reduction core process can be avoided by using hydrogen instead of natural gas (see also Figure 8).
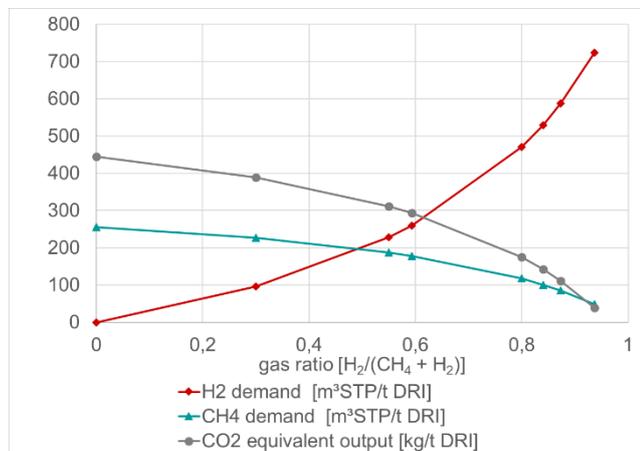
Figure 8: Direct CO_2-emissions of DR-NG and DR-H_2 process

For a complete examination of the carbon footprint of DR-processes also the indirect CO_2 emissions associated with the required electricity have to be considered. Especially for the DR-H_2 process the carbon intensity of the electricity used for the generation of hydrogen via electrolysis is a main influencing factor on total CO_2-emissions. compared to the input of natural gas. That means, that the main carbon sink for the hydrogen based direct reduction process is the carbon fraction of the produced DRI.

Figure 9 presents an overview of total CO_2 equivalent output depending on the amount of hydrogen used in the process and CO_2-intensity of electricity production. The higher the share of hydrogen in the reducing gas, the lower the absolute CO_2-emissions but also compared to the input of natural gas. That means, that the main carbon sink for the hydrogen based direct reduction process is the carbon fraction of the produced DRI.
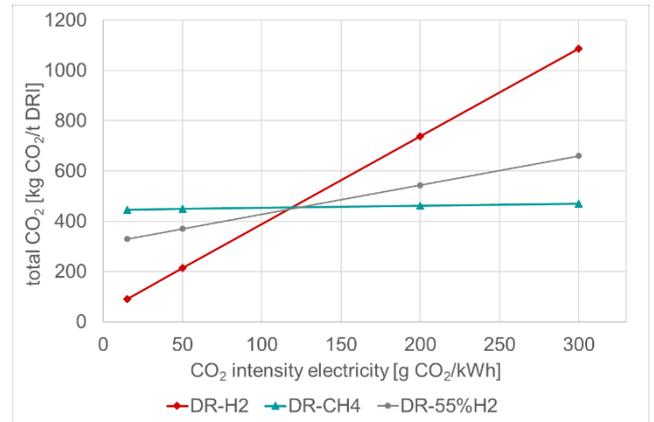
Figure 9: Dependency of total CO_2-emissions on CO_2-intensity of electricity

Conclusion and outlook

The potential for CO_2-savings with the current steel production routes is very limitied and is far away to achieve the 2050`s climate goals. The present reseach shows that the implementation of the DRI-H_2 process route can unlock a huge potential for the reduction of CO_2 emissions from steelmaking. The evaluation of the process shows significant decrease on the CO_2. When compared to the DR-NG, the achievable CO_2-saving of direct emissions is 91%. Nevertheless, as reported in this paper, the total CO_2 reduction depends strongly on the carbon intensity of the supplied electricity and will be only feasible, when the system fully operates with renewable energy. This means that one of the premises for greening the iron and steel industry is the decarbonization of the electricity sector.

Apart from the technological feasibility of operating a direct reduction plant with hydrogen, further questions arise that can only be partially answered by the steel industry.

The calculated electricity needs to produce hydrogen was about 3,500 kWh/t DRI. When the hydrogen production is enlarged at European level the annual energy needs will increase up to 400-500 TWh per year. An important increase, considering that the actual electricity consumption of the steel industry is rated on 75 TWh [2].

Furthermore, for the hydrogen based operation of a direct reduction plant like the one of voestalpine in Texas with 2 million t DRI per year, an electrolysis plant in the GW range would be required to supply

green hydrogen. Electrolysers of this size are currently not available on the market, the largest PEM electrolysis plants under construction are currently reaching the double-digit MW range. The availability, maintenance and associated operating costs of these plants in an industrial environment are still difficult for potential operators to assess and is to be found, for example in the context of the funded project H2FUTURE by operating a PEM electrolysis plant with a nominal power of 6 MW.

Furthermore, the supply of electricity for the operation of large-scale hydrogen-based reduction plants must be continuous throughout the year. Therefore, in addition to the technical questions on electrolysis technology, the transfer of energy systems away from fossil to CO_2-free generation is a prerequisite. Shifting to a sustainable steelmaking in the EU, an additional requirement for renewable electrical energy of about 18 % of the current EU total consumption is necessary [2]. This poses a huge challenge to all stakeholders involved.

Abbreviations

BF-BOF	Blast furnace/Basic oxygen furnace
DR	Direct reduction
DR-H_2	Direct reduction with hydrogen
DRI	Direct reduced iron
DR-NG	Direct reduction with natural gas
EAF	Electric arc furnace
HBI	Hot briquetted iron
PEM	Proton Exchange Membrane
STP	Standard conditions (0°C, 1013 mbar)

Acknowledgments

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