

# Green Hydrogen-Based Direct Reduction for Low-Carbon Steelmaking

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The European steel industry aims at a CO<sub>2</sub> reduction of 80-95% by 2050, ensuring that Europe will meet the requirements of the Paris Agreement. As the reduction potentials of the current steelmaking routes are low, the transfer toward breakthrough-technologies is essential to reach these goals. Hydrogenbased steelmaking is one approach to realize CO2-lean steelmaking. Therefore, the natural gas (NG)-based direct reduction (DR) acts as a basis for the first step of this transition. The high flexibility of this route allows the gradual addition of hydrogen and, in a long-term view, runs the process with pure hydrogen. Modelbased calculations are performed to assess the possibilities for injecting hydrogen. Therefore, NG- and hydrogen-based DR models are developed to create new process know-how and enable an evaluation of these processes in terms of energy demand, CO2-reduction potentials, and so on. The examinations show that the hydrogen-based route offers a huge potential for green steelmaking which is strongly depending on the carbon footprint of the electricity used for the production of hydrogen. Only if the carbon intensity is less than about 120 g  $CO_2$  kWh<sup>-1</sup>, the hydrogen-based process emits less CO<sub>2</sub> than the NG-based DR process.

# 1. Introduction

The iron and steel industry as one of the biggest industrial emitters of  $CO_2$  accounts for approximately 30% of the global industrial  $CO_2$  emissions. As set in the Paris Agreement, the emissions of greenhouse gases have to be reduced by 80–95% until 2050 compared with preindustrial levels to keep global warming below 2 °C.<sup>[1]</sup>

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Currently, crude steel production in the European Union is almost entirely divided between the blast furnace/basic oxygen furnace (BF/BOF) route and the scrap-based electric arc furnace (EAF) route. In 2018, 58.3% are produced via the BF/BOF route, whereas 41.7% of the crude steel is produced via the EAF route.<sup>[2]</sup> To reach the goal of 80% CO<sub>2</sub> reduction, it is indispensable to implement so-called "breakthroughtechnologies" in future steelmaking systems. Since currently applied steelmaking processes are already operating close to their thermodynamic limits, the CO<sub>2</sub>-abatement potential of these routes is limited. Regarding the most prevalent route, the BF/BOF route, about 1.9 t of CO<sub>2</sub> are emitted per ton of crude steel on average, including agglomeration, iron- and steelmaking, casting, and hot rolling.<sup>[3]</sup> The maximum CO2-reduction potential attainable with the actual production routes

(considering the decrease in  $CO_2$  intensity of the power sector as well as the increase in scrap availability) is predicted to be about 15% between 2010 and 2050.<sup>[4]</sup> Due to the lower carbon footprint of the scrap-based EAF route, this process is clearly in advantage regarding  $CO_2$  emissions (455 kg  $CO_2 t^{-1}_{CS}$ ).<sup>[3]</sup> However, natural iron sources as raw material will still be necessary in future due to limited availability of scrap and required steel qualities, even though the forecasts for rising scrap availabilities in future are slightly higher than that for total steel production (1.1% annual increase for scrap availability vs 0.5% growth rate for European steel production).<sup>[4]</sup>

For achieving substantial  $CO_2$ -reduction rates in the steel sector, two main approaches can be distinguished: 1) Smart Carbon Usage (SCU) including process integrated measures for a decreased use of carbon in existing processes and utilization of  $CO_2$  as a raw material for chemical conversion (carbon capture and utilization), optionally combined with carbon capture and storage (CCS). 2) Replacement of carbon by renewable electricity and/or fossil-free reductants to directly avoid  $CO_2$  emissions (Carbon Direct Avoidance [CDA]).<sup>[4]</sup>

CDA processes can mainly be split into hydrogen- and electricity-based reduction processes. Electrical power-based iron reduction technologies use electricity to produce steel by means of iron ore electrolysis at different temperature levels (low-temperature iron electrowinning, high-temperature pyroelectrolysis).<sup>[5–7]</sup> They provide high potentials for CO<sub>2</sub>

reduction of up to 95% if 100% renewable electricity is used.<sup>[8]</sup> These processes are currently under development.

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The same also applies for processes replacing carboncontaining reducing agents by hydrogen either in solid state (direct reduction [DR]) or in liquid state (plasma smelting reduction). Hydrogen plasma smelting reduction directly converts iron ore fines to liquid steel via hydrogen in ionized form. Hydrogen plasma is used for the reduction of the oxides and simultaneously provides heat for melting the metallic iron.<sup>[9]</sup> DR processes with natural gas (DR-NG) as one of the state-of-the-art steelmaking technologies can provide the basis for the introduction of hydrogen. Existing DR-NG processes already operate with a hydrogen containing syngas, which is previously produced by the reforming of NG. Based on this process concept, additional amounts of hydrogen can be implemented to achieve a further CO<sub>2</sub> reduction. However, there is little information about the behavior of hydrogen-based DR processes available in the literature. Therefore, the scope of this work is to present potential designs of a DR process concept based on hydrogen to gain detailed process know-how about the behavior of hydrogen in DR processes. Advances in research are achieved with reference to a direct comparison of NG- and hydrogen-based iron ore reduction to evaluate these processes in terms of energy demand, type of electricity used for steel production, CO2-reduction potentials, and so on.

# 2. Current State of NG- and Hydrogen-Based DR Processes

DR processes together with an electric arc furnace provide the basis for CO<sub>2</sub> reduction in the steel industry. In case of using NG as reducing agent approximately one-third of the CO<sub>2</sub> emissions can be saved compared with the BF/BOF route.<sup>[10]</sup> These savings are achieved due to the higher hydrogen content from NG which also acts as reducing agent. Currently, there are two main technology providers for NG-based DR processes:

HYL/Energiron and MIDREX. Due to its high implementation rate, representing about 65% of the total worldwide produced direct reduced iron (DRI),<sup>[11]</sup> the MIDREX process was selected as basis for the following considerations (see **Figure 1**). The key component of the DR process is a shaft furnace, where the reduction of iron ore to sponge iron by using NG or other gaseous reducing agents takes place.

Lump ore or pellets are charged from the top of the shaft furnace as raw material and reduced inside the furnace producing direct reduced iron according to the following overall reduction reactions<sup>[13]</sup>

 $Fe_2O_3 + 3H_2 \rightarrow 2Fe + 3H_2O \ \Delta H_R = 99 \, kJ \, mol^{-1}$  (1)

$$Fe_2O_3 + 3CO \rightarrow 2Fe + 3CO_2 \ \Delta H_R = -24 \text{ kJ mol}^{-1}$$
 (2)

The oxygen from the iron ore reacts with the reducing gas, mainly composed by a mixture of CO and  $H_2$  at elevated temperatures to produce metallic iron while releasing CO2 and H<sub>2</sub>O. The heat required for the endothermic reduction with hydrogen is supplied by the energy released from the exothermic reaction between carbon monoxide and iron oxides. The reducing gas (CO and H<sub>2</sub>) is fed in the middle section of the shaft furnace and thus reduces the ferrous material. The exhaust gas from the reduction-the so-called top gasprincipally comprises CO<sub>2</sub> and H<sub>2</sub>O. It is cleaned and cooled in a top gas scrubber, where water is partly condensed and the dust is removed, and then recirculated for reuse. Once blended with NG, about two-thirds of those gases return as feed to the reformer.<sup>[14]</sup> The remaining part is used as a heating source for the reformer, where it is combusted with air. The mixture of NG and top gas passes through the reformer pipes, which are filled with nickel catalyst. Thus, a reducing gas with a composition of about 55% of hydrogen and 35% of carbon monoxide is produced according to the reactions given in Equation (3) and (4). Steam methane and dry reforming reactions



Figure 1. MIDREX-DR process with possible hydrogen addition.<sup>[12]</sup>

take place simultaneously in the reformer at 900–950  $^\circ C.^{[15]}$  Therefore, parts of the CO<sub>2</sub> are recycled to provide CO for the reduction of iron oxides.^[16]

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$$CH_4 + CO_2 \rightarrow 2CO + 2H_2 \ \Delta H_R = 247 \, \text{kJ} \, \text{mol}^{-1} \tag{3}$$

$$CH_4 + H_2O \rightarrow CO + 3H_2 \quad \Delta H_R = 206 \text{ kJ mol}^{-1}$$
(4)

An alternative pathway to achieve a further reduction of  $CO_2$  emissions is the utilization of renewable hydrogen as energy source and reducing agent for the production of DRI. As the reforming gases in the DR process already comprise 55% hydrogen, NG can be partly replaced by hydrogen and thus achieve a further reduction in  $CO_2$  emissions. As stated by Ripke et al.,<sup>[12]</sup> no changes are required for the existing processing plants up to a substitution rate of about one-third of the required NG.

Using  $H_2$  as reducing agent enables achieving higher reduction degrees from iron ore. Nevertheless, the reduction process is thermally unfavorable due to the endothermic nature of the reaction between hydrogen and iron oxide.<sup>[17]</sup>

Hydrogen could be used as well as single reducing agent in the DR process. In this case, the top gas is principally composed of water. No reformer would be required. Instead, a gas heater will be attached to the system to preheat the gases to the required temperatures. Either hydrogen or other environmentally friendly heat sources (NG, green electricity, waste heat) might be used as fuels for the heater.<sup>[12]</sup>

According to MIDREX, 800 m<sup>3</sup><sub>STP</sub> t<sup>-1</sup><sub>DRI</sub> of hydrogen would be necessary for the full operation with hydrogen. The reduction process itself would need 550 m<sup>3</sup><sub>STP</sub> t<sup>-1</sup><sub>DRI</sub>, whereas 250 m<sup>3</sup><sub>STP</sub> t<sup>-1</sup><sub>DRI</sub> of hydrogen would be required as fuel for the gas heater. Furthermore, about 50 m<sup>3</sup><sub>STP</sub> t<sup>-1</sup><sub>DRI</sub> of NG have to be added to maintain the temperature and carbon content of the DRI.<sup>[18]</sup>

The temperature decrease resulting from the endothermic hydrogen reduction reaction in the shaft furnace can also be compensated by the addition of NG. Thus, energy is released as a consequence of the exothermic reaction between  $Fe_2O_3$  and CO. In addition, NG aids to maintain the desired carbon content in the DRI, as seen in the carburization reactions described as follows<sup>[12]</sup>

$$3Fe + CO + H_2 \rightarrow Fe_3C + H_2O \tag{5}$$

 $3Fe + CH_4 \rightarrow Fe_3C + 2H_2 \tag{6}$ 

$$3Fe + 2CO \rightarrow Fe_3C + CO_2$$
 (7)

Three reasons make the DRI carbon content critical when used in an electric arc furnace: 1) The presence of carbon is necessary to complete the metallization of the iron in the EAF. 2) Carbon represents an additional source of energy in the EAF because burning the carbon by injecting oxygen reduces the electricity consumption, consequently enabling a faster melting of the charged materials. 3) Carbon enables the formation of a foamy slag in the EAF.<sup>[15]</sup> The preferred optimum carbon content in the DRI is about 1.5–3% despite it deviates significantly with the material input and the produced steel grades. Ripke et al. reported that a DRI carbon content of about 1.4% could be reached if 50  $m_{STP}^3 t^{-1}_{DRI}$  of NG were added.<sup>[12]</sup>

Due to varied possibilities for the implementation of hydrogen in the DR process and distinctions in process concepts (e.g., configuration for heaters), different information about the hydrogen demand for a fully hydrogen operated DR process can be found in the literature. Although the available information is very limited, the following values present an overview thereof. Vogl et al.<sup>[19]</sup> reported a hydrogen demand of 51 kg t<sup>-1</sup> of steel, equivalent to about 520 m<sup>3</sup><sub>STP</sub> t<sup>-1</sup> of hot briquetted iron (HBI). According to their description, the model consisted of a DR process operated by hydrogen and combined with an EAF. As the reducing gas was heated up by the recovered heat from the condenser, no additional heating source was required. In the model, the additional energy for the preheating the iron ore was as well taken into account.

Simulations performed by ArcelorMittal Hamburg GmbH owner of the single DR-plant in Europe with an annual production of approximately 0.55 Mt<sup>-1</sup> of cold DRI—showed an overall hydrogen demand of 635  $m_{STP}^3 t_{HBI}^{-1}$  including energy losses. Regarding the energy requirements, 3 MWh  $t_{HBI}^{-1}$  were reported for the hydrogen generation and additional 0.23 MWh<sub>el</sub>  $t_{HBI}^{-1}$  for the electrical heating of the reducing gas.<sup>[20]</sup>

# 3. Modeling Approach

Within the scope of the current work, process models are developed to evaluate hydrogen-based DR concepts. Furthermore, these developed models are compared with the NG-based technology with additional injection of hydrogen. The models are set up in an equation-oriented process simulation platform using the m.SIMTOP model library for metallurgical processes which has been developed by Primetals Technologies and voestalpine in recent years.<sup>[21]</sup> Process configurations presented in refs. [12,18] provide the basis for the chosen layout, as voestalpine is also running a MIDREX DR-plant in Corpus Christi, Texas, since 2017.<sup>[22]</sup> The goal of the model development efforts is to identify carbon streams in the DR process and to assess CO<sub>2</sub>-abatement potentials as well as energy flows.

The starting point of the current work is to set up a model for a state-of-the-art DR-NG process which acts as a reference case for further considerations. Based on this case, opportunities for injecting additional hydrogen into the process are implemented. The layout of this model including its corresponding system boundaries is shown in **Figure 2**. As the focus of the research work is the evaluation and comparison of different reducing agents (NG, H<sub>2</sub>) of the DR process, no up- or downstream processes such as a pelletizing plant or an electric arc furnace and their corresponding emissions or energy demands are included in the calculations. The same also applies for input streams like NG or lump ore whose emissions from mining and transport are not included.

The main unit operations of the process comprise DR-shaft, reformer, top gas scrubber, product gas compressors, and heat exchangers. In addition, several material sources and sinks as well as gas splitters and mixers are included. **Table 1** shows an overview of the most relevant process parameters of the developed models.

Iron input materials like pellets and lump ore are introduced into the processes by using ore source models. Additives like



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Figure 2. Process model for (NG-based) DR-NG process with addition of hydrogen.

Table 1. Main modeling parameters.

Description	Unit	DR-NG process	DR-H <sub>2</sub> process
Iron input material	$kg t^{-1}_{DRI}$	1.391	1.391
DRI temperature	°C	730	730
Reduction degree	%	95	95
Carbon content of DRI	%	1.7	1.7
Rist diagram pinch point (FeO/Fe reduction equilibrium)	°C	820	820
Bustle gas temperature	°C	887–908	980
Bustle gas oxidation degree	%	9.25–15.6	10.25
Product gas temperature (scrubber outlet)	°C	70	40
Top gas fuel temperature (scrubber outlet)	°C	45	40

lime are supplied by a separate additive source model, whereas gaseous inputs such as NG, hydrogen, or seal gas are implemented through gas source models. The main output streams comprise hot direct reduced iron at a temperature level of approximately 700 °C as well as off gas of the reformer.

The shaft reactor itself, which is the main component for the reduction process, is simulated as a two-zone model. This model includes heating and reduction zones representing the reduction reactions given in Equation (3)–(7). The counter current flow regime is established with the reduction gas entering the shaft model at the bottom and the solid input stream entering at the top of the shaft. This gas phase ascends through the reaction zones, where it heats up and reacts with the solid input stream which descends to the bottom. The assumed reduction degree for

the process model is set to 95%. A target value of 1.7% for the carbon content of the produced DRI is set.

The main gas and solid streams are connected to a submodel which is used to analyze and graphically illustrate the thermodynamic boundary conditions of the reduction process in the form of a so-called Rist diagram. The Rist diagram has been developed since the 1960s as graphical representation of the blast furnace process.<sup>[23]</sup> Furthermore, it also has been demonstrated to be an effective tool to evaluate the process conditions of indirect reduction processes and enables the determination of the gas utilization. As shown in Figure 3, the Rist diagram can be adapted to illustrate the thermodynamic characteristics of the DR process. In this modified form, the Y-axis depicts the reduction degree of iron oxides in the solid material, whereas the X-axis is used to describe the oxidation degree of the reduction gas phase. The operation line of the diagram is characteristic for the countercurrent oxygen exchange process in the shaft furnace. The low end point of this operation line is equivalent to DRI with a reduction degree of  $Y_R$ , which is produced using reduction gas with the initial oxidation degree  $X_{\rm R}$ . The high end point of the operation line is representative for solid input material in the form of hematite (Y = 1.5) and the final oxidation degree of the reduction gas after leaving the shaft furnace.<sup>[24]</sup> The equilibrium line in the diagram is based on a projection of the reduction equilibria (M, W) of magnetite (Fe<sub>3</sub>O<sub>4</sub>) to wustite (FeO) and wustite (FeO) to metallic iron (Fe). This projection is dependent on the characteristic reduction temperature in the shaft furnace ( $\vartheta_{BG}$ ) and the initial reduction gas composition. The thermodynamic efficiency of the reduction process corresponds to the approach of the operation line to the wustite corner (W) of the equilibrium line.<sup>[25]</sup> It can be viewed as the thermodynamic pinch point of the reduction process.



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Figure 3. Example of a Rist operating diagram to depict a DR process, adapted with permission from ref. [24,25].

The reformer of the DR-NG model comprises one part for the reforming of NG following the chemical reactions (3) and (4) as well as a heating section for providing the required energy for producing the syngas. The reforming process approaches a state of thermodynamic equilibrium in practice. To consider this behavior, minor amounts of methane are bypassed and thus excluded from the chemical reactions of the reforming process. The necessary energy is provided through a gas combustion submodel.

The third main component is the top gas scrubber which has the goal to reduce the water content on the one hand and control the temperature levels of the two separated gas output streams on the other hand. Therefore, the temperature and the pressure level of these two streams—reducing gas input and top gas fuel—are set to fixed values. Each gas stream is assumed to reach a state of saturation which is estimated according to the so-called Antoine formulations. NG 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. The input of hydrogen in the DR-NG process model is foreseen in the reforming gas before entering the reformer.

As already mentioned, some process steps have to be adapted for running the DR process with hydrogen as single reducing agent. Therefore, a second model (DR-H<sub>2</sub>), which is shown in **Figure 4**, was developed. The reformer of the DR-NG process is replaced by a reduction gas heater representing a combination of a gas burner, mixer, and heat exchanger model. Hydrogen can be added either to the reduction gas and/or to the top gas fuel for heating purposes. The energy supplied to the heater can also be provided by NG.

To achieve comparable results, the same solid input material compositions and amounts as well as the same basic assumptions for the DR shaft (e.g., reduction degree, carburization behavior, temperature distribution, and so on), the top gas scrubber (temperatures and pressure levels) and the gas burner (excess air) are applied for both process models.

## 4. Results and Discussion

The two different models of DR processes proposed in the previous section are analyzed in terms of reducing agent and energy demands, CO<sub>2</sub>-abatement potentials, and so on. These results are presented subsequently. Starting from the DR-NG reference process operating with NG as the sole reducing agent, the implementation of hydrogen is evaluated for both the gradual injection of hydrogen to the NG-based system and the H<sub>2</sub>-DR process



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Figure 4. Process model for (hydrogen-based) DR-H<sub>2</sub> process.

working with pure hydrogen. The current process models were slightly adapted compared with the models presented at the ESTAD conference<sup>[26]</sup> in terms of an optimized control strategy as well as a further developed modeling approach for several unit operations: in the presented model, two separate temperature and pressure levels were applied to the resulting gas output streams of the top gas scrubber. In addition to this, the thermodynamic boundary conditions of the reduction process in the shaft were derived from a Rist diagram submodel (as described in Section 3) and taken into consideration for the implementation of hydrogen.

#### 4.1. DR-NG Process with Hydrogen Addition

To reach a reduction degree of the hot DRI product of 95%, 1.4 t of iron oxide input material containing input materials per ton of DRI are required for the DR-NG as well as the DR-H<sub>2</sub> process. Based on NG which contains approximately 95% methane, the simulation results in an NG demand of approximately 259 m<sup>3</sup><sub>STP</sub> t<sup>-1</sup><sub>DRI</sub> for the reference case. This amount corresponds with the given data from the literature which are about 262 m<sup>3</sup><sub>STP</sub> t<sup>-1</sup><sub>DRI</sub>.<sup>[27]</sup>

Based on this reference case, a gradual injection of hydrogen was implemented to the model (see **Figure 5**). Following the information given by Ripke et al.,<sup>[12]</sup> about 30% of the NG can be replaced by hydrogen without any process changes. This data, among others, was used in the DR-NG model and resulted in a reduction of NG demand by about 80  $\text{m}^3_{\text{STP}} \text{t}^{-1}_{\text{DRI}}$  and a corresponding hydrogen demand of 270  $\text{m}^3_{\text{STP}} \text{t}^{-1}_{\text{DRI}}$ . In principle, the model also compiled for higher H<sub>2</sub>/(H<sub>2</sub> + CH<sub>4</sub>) volume ratios up to 0.85. However, for an increased addition



Figure 5. Reduction gas demand for DR-NG process including hydrogen addition.

of hydrogen, various factors that assert influence on the operation of the DR process have to be considered.

#### 4.1.1. Impact of Feeding Hydrogen

As shown in **Figure 6**, the replacement of NG by hydrogen leads to an increased volume flow in the shaft furnace due to changing gas compositions. The gradual injection of hydrogen entails a corresponding decrease in carbon monoxide, whereas the content of  $CH_4$  is only slightly influenced by higher hydrogen shares. This behavior can be explained by the fact that even if 100% of NG is used, it is almost completely decomposed to carbon monoxide and hydrogen in the reformer before entering the shaft. Only a small and constant amount of NG is added for the enrichment of the bustle gas before entering the shaft.





Figure 6. Compositions and flow of shaft gas.

Due to the increased gas amounts, different process conditions in the shaft furnace have to be considered. Rising gas velocities will lead to a higher pressure drop in the shaft furnace. Thus, different gas residence times and operating conditions could affect the chemical reactions taking place in the furnace. Moreover, gas compositions and temperatures as well as the metallization degree could be affected because of the changing reducing agents.

As the gas composition considerably influences the reduction rate, the reduction potentials of the different components have to be considered. For example, the reduction potential of hydrogen is higher than that of carbon monoxide at the given temperature levels of about 900 °C.<sup>[28]</sup>

Regarding the water gas shift reaction (Equation (8)),<sup>[29]</sup> the equilibrium will be shifted to the left. Thus, the generation of CO is favored which may further improve the carburization reactions given in Equation (5)–(7). In addition, hydrogen shows an improved kinetic reduction behavior compared with carbon monoxide.<sup>[28]</sup>

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

In addition to the changed process conditions in the shaft furnace, also other unit operations of the process chain could be affected by varying gas compositions and amounts. Due to the decreased methane consumption, the overall heat duty in the reformer declines. To ensure a constant reformed gas temperature, the amount of cleaned top gas which is directed to the gas burner of the reformer has to be reduced. Otherwise, the reformer flue gas temperature would increase. Furthermore, additional water can be removed from the reforming gas outlet stream in the scrubber to compensate for the increased gas amounts. This will require additional cooling capacity to be installed. The potential effects of the decreased water content of the reducing gas in the reformer with respect to coke formation have to be examined.

In general, hydrogen can be introduced at different stages of the process chain. Thus, operating conditions in the reformer and required heat amounts could be affected. For example, hydrogen can be added either right before entering the shaft furnace as it is shown in Figure 1 or to the process gas before entering the reformer. If hydrogen is added directly before the shaft furnace, it has to be preheated in a separate heating unit. As shown in Figure 2, hydrogen is injected before the reformer in the presented model so that an additional heating unit can be avoided. However, the kinetics of the reforming reactions (Equation (3) and (4)) could be decreased because of higher hydrogen shares in the reformer pipes.

#### 4.2. Hydrogen-Based DR Process (DR-H<sub>2</sub>)

The second model (DR-H<sub>2</sub>) was calculated for an input of about 95% hydrogen. The remaining part is NG which is necessary to maintain process temperatures and the carbon content of the produced DRI. Process parameters and boundary conditions such as the amount and compositions of iron containing input materials, DRI temperature and composition, and so on were kept constant compared with the reference DR-NG model.

Depending on the assumed hydrogen content of the bustle gas, 726–767  $\rm m^3_{STP} t^{-1}_{DRI}$  of hydrogen are calculated for the hydrogen-based DR model. This data slightly deviates from the values given in ref. [18] which are approximately 800  $\rm m^3_{STP} t^{-1}_{DRI}$ . These deviations could originate from different boundary conditions applied for the models and the consideration of losses. For example, no losses such as seal gas losses or others, which will lead to higher reducing gas and accordingly hydrogen demands, are included in the presented model. In addition to the hydrogen, also 48  $\rm m^3_{STP} t^{-1}_{DRI}$  of NG are required as input to the system.

Hydrogen is needed at two different process stages in the considered model. For the reduction process between 484 and 525  $m_{STP}^3 t_{DRI}^{-1}$  are required. These amounts correspond with the data reported by Vogl et al. (520  $m_{STP}^3 t_{HBI}^{-1}$ ).<sup>[19]</sup> According to their process concept, hydrogen was only used for the reduction of iron oxides.

The remaining part is used to compensate for temperature losses arising from the endothermic nature of the hydrogen reduction reaction (Equation (1)) on the one hand. On the other hand, sensible heat for heating up the reduction gas has to be supplied. As described earlier, this amount of hydrogen could also be replaced by alternative energy sources. The effect of the different heating sources on the overall energy balance has to be further investigated.

#### 4.2.1. Energy Demand

As the hydrogen-based DR process is operated almost without the utilization of carbonaceous fossil sources, the required energy input to the process has to be provided by alternative sources. In the case of hydrogen, it has to be generated from renewable energy to obtain a CO<sub>2</sub>-lean steel production. Thus, the electrolysis of hydrogen is currently the most promising technology for a green hydrogen production. An additional electricity demand of approximately 3400–3600 kWh t<sup>-1</sup><sub>DRI</sub> is required for the production of hydrogen considering electrolysis with an efficiency of 75% based on the higher heating value of hydrogen (3.54 kWh m<sup>-3</sup><sub>STP</sub>). This data includes the generation of the reducing gas as well as the hydrogen demand for heating the reducing gas. If the hydrogen demand for heating is provided by waste heat from other processes or other alternative



energy sources, an increased efficiency can be reached. This will result in a remaining minimum energy demand of 2300–2500 kWh t<sup>-1</sup><sub>DRI</sub> which is required for the reduction of iron ores itself. In addition, approximately 80–125 kWh t<sup>-1</sup><sub>DRI</sub> are necessary to cover the electricity demand for auxiliaries such as compressors, water supply, and others.<sup>[10,20]</sup> For an evaluation of the entire steel production chain, also energy demands of upstream processes and particularly of the EAF have to be considered. Thus, operating an EAF requires an additional amount in the range of 310–640 kWh t<sup>-1</sup><sub>CS</sub> depending on the share of scrap/DRI (increasing DRI amounts lead to higher energy demands), temperature of DRI, specific slag mass, and so on.<sup>[30]</sup>

#### 4.3. CO<sub>2</sub>-Reduction Potentials

The main goal to apply hydrogen for DR processes is to achieve a substantial reduction of  $CO_2$  emissions in future steelmaking. Based on the previous presented results, the hydrogen-based DR processes are evaluated regarding their  $CO_2$ -mitigation potentials. Therefore, carbon input and output streams as well as the distribution of carbon flows within the process are quantified for both process models (see **Figure 7** and **8**).

NG either used for reforming, enrichment, or heating purposes represents the main carbon input to the DR-NG reference process. In addition to the carbon output via the DRI product, the



Figure 7. Gas  $[m_{STP}^3 t^{-1} _{DRI}]$ , carbon [kg C  $t^{-1} _{DRI}]$ , and water [t H<sub>2</sub>O  $t^{-1} _{DRI}]$  flows for DR-NG process.



**Figure 8.** Gas  $[m_{STP}^3 t^{-1} _{DRi}]$ , carbon [kg C  $t_{DRi}^{-1}$ ], and water [t H<sub>2</sub>O  $t_{DRi}^{-1}$ ] flows for DR-H<sub>2</sub> process.



major emission source of carbon is the off gas of the reformer. About 124 kg C t<sup>-1</sup><sub>DRI</sub> which are equivalent to about 453 kg  $CO_2 t^{-1}_{DRI}$  are emitted at this point. In comparison, the carbon output of the DR-H<sub>2</sub> process is almost equally distributed between the DRI and stack emissions (released by using the top gas as combustion gas for the heater), representing 17 and 11 kg C t<sup>-1</sup><sub>DRI</sub>, respectively. The main carbon source in this case is as well NG which is required for maintaining the carbon content of the DRI. Even though this carbon does not implicate any CO<sub>2</sub> emissions in the DR process, it has to be considered that CO<sub>2</sub> is released during the melting and refining in the EAF. Direct CO<sub>2</sub> emissions in the range of 72–180 kg CO<sub>2</sub> t<sup>-1</sup><sub>CS</sub> are generated from electric arc furnaces.<sup>[31]</sup>

According to the DR-H<sub>2</sub> process setup, higher recirculating gas flows are necessary to maintain the required process temperatures. Residual amounts of CO and CO<sub>2</sub> are still present in the recycled gas and increase the total specific volume flow in the system. However, carbon containing gas streams only play a minor role in the hydrogen-based DR process.

To get an idea how the transition from the utilization of NG as reducing agent toward hydrogen influences the  $CO_2$  balance, **Figure 9** shows the corresponding decrease in  $CO_2$  formation with rising H<sub>2</sub>-input ratios. Starting from a directly emitted  $CO_2$  amount of 453 kg  $CO_2 t^{-1}_{DRI}$  for the NG-reference case, the emissions can be reduced to a level of 40 kg  $CO_2 t^{-1}_{DRI}$  for the DR-H<sub>2</sub> process. Thus, up to 91% of direct  $CO_2$  emissions of the DR core process can be avoided by using hydrogen instead of NG.

To get a complete overview of the carbon footprint of the DR process, not only the direct emissions of the process itself have to be regarded, but also indirect emissions from the generation of the used electricity must be taken into consideration. Taking electrolysis as production technology for hydrogen into account, the carbon intensity of the provided electricity mainly influences the CO<sub>2</sub> footprint of the whole DR-H<sub>2</sub> production process. The influences of CO<sub>2</sub> intensity of electricity on the complete DR process (based on NG, H<sub>2</sub>, and a mixture of NG and H<sub>2</sub>) are shown in **Figure 10**. The DR-NG reference process is nearly independent from the CO<sub>2</sub> footprint of the electricity, whereas the dependency increases with rising shares of hydrogen input. The calculations show that the "break-even-point" for the CO<sub>2</sub> intensity of the electricity for hydrogen-based DR is about



Figure 9. Direct  $CO_2$  emissions of DR-NG and DR-H<sub>2</sub> process.



Figure 10. Dependency of total  $CO_2$  emissions of DR process on  $CO_2$  intensity of electricity.

 $120 \text{ g CO}_2 \text{ kWh}^{-1}$ . This means that only if the CO<sub>2</sub> intensity is below that value the carbon footprint of the H<sub>2</sub>-DR process is lower than that for the NG-based route.

# 5. Conclusion

Achieving the 2050's climate goals seems to be unattainable with currently applied steel production routes. As hydrogen-based DR of iron ores is one of the breakthrough technologies to enable CO2-lean steel production, a detailed examination thereof was conducted within the scope of the current work. The evaluation of the process shows significant potentials for the reduction of CO<sub>2</sub>, if hydrogen is used as reducing agent. Compared with the DR-NG reference process up to 91% of directly emitted CO<sub>2</sub> emissions can be reduced without taking the CO<sub>2</sub> intensity of the additionally required electricity into account. However, the overall CO2 footprint especially for hydrogen-based DR is strongly depending on them due to the production of hydrogen via electrolysis. Only if the system fully operates with renewable electricity, the goals for a substantial CO<sub>2</sub> abatement can be reached. This implicates a deep decarbonization of the electricity sector as one of the prerequisites for green steelmaking. For a further evaluation of the overall steelmaking process, the boundaries of the regarded process steps have to be expanded. Thus, also CO<sub>2</sub> emissions from up- and downstream processes or input materials like the pelletizing process, upstream emissions of NG, and main downstream processes such as the electric arc furnace have to be considered.

Apart from the CO<sub>2</sub>-reduction potentials, some technological questions concerning the feasibility of operating a DR plant with hydrogen such as the reaction behavior in the shaft furnace or influences on different unit operations like the top gas scrubber have to be solved. Furthermore, the continuous supply and production of hydrogen on industrial scale are essential for the realization of a sustainable steelmaking process. For example, running a DR plant with an annual production capacity of about 2 million t DRI year<sup>-1</sup> (like the one of voestalpine in Texas) with hydrogen, the required capacity of the electrolyzer is in the GW range. Currently, the largest proton exchange membrane (PEM) electrolysis plants under construction reach the double-digit MW range. This difference implicates high efforts



for scaling-up and further developing existing electrolyzer technologies. The availability, maintenance, and associated operating costs of these plants in an industrial environment are still difficult for potential operators to evaluate. By operating a PEM electrolysis plant with a nominal power of 6 MW, these issues are examined within the context of the funded project H2FUTURE.<sup>[32]</sup>

In addition to the technical questions concerning the electrolysis technology, the transfer of the energy systems toward renewables has to be realized. A continuous supply of renewable electricity throughout the year has to be ensured for the operation of large-scale hydrogen-based DR plants. Shifting to a CO<sub>2</sub>-lean steelmaking in the European Union, the demand of renewable electricity will rise drastically in future, considering the fact that about 3500 kWh of electricity are required for the hydrogenbased production of 1t DRI. Rolling the required energy amounts of low-carbon steel production out on a European level, an additional annual energy demand of 400-500 TWh arises. An important increase, considering that the actual electricity consumption of the steel industry, is rated on 75 TWh and that the additional energy requirements corresponds to 18% of the current EU total consumption.<sup>[8]</sup> Even if the additional energy demand would be lower due to the application of other lowcarbon or scrap-based steelmaking routes, for example, the development of suitable electrolyzers and the supply of sufficient amount of renewable energy will be a huge challenge for all involved stakeholders, which has to be tackled in future.

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# **Conflict of Interest**

The authors declare no conflict of interest.

# **Keywords**

CO2 emissions, direct reduction, energy demand, hydrogen, natural gas

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