

CO₂ Emission Reduction Potential in the Steel Industry by Integration of a Direct Reduction Process into Existing Steel Mills

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Abstract

In the context of climate change, the reduction of greenhouse gas emissions in all economic sectors is considered to be an important factor in order to meet the demands of a sustainable energy system. The steel industry as one of the large industrial CO₂ emitters is currently highly dependent on fossil resources. In order to reduce coke consumption and thereby CO₂ emissions while still being able to further utilize existing blast furnaces, the possibility of including a direct reduction process (DRP) into a fully integrated steel mill was investigated. Therefore, a blast furnace model, derived from literature data and implemented in Aspen Plus, was used to analyze the impact of DRI in the blast furnace process. Furthermore, a state-of-the-art DRP was modeled to investigate the possibility of substituting the reducing agent natural gas with hydrogen. A sensitivity analysis was carried out in order to find the boundary percentage of hydrogen as a reducing agent without penalty to the DRI quality. Lastly, the two modeled process steps were combined to form a route of producing hot metal. By varying boundary conditions of the DRP while recording the CO₂ emissions of the two process steps, the overall potential for the reduction of CO₂ emissions was estimated. Within the simulated range a maximum reduction of CO₂ emissions of 25.8% relative to typical emissions of a blast furnace could be determined.

Introduction

Since the 195 member states of the Paris agreement concluded that global temperature rise has to be kept well below 2 °C compared to pre-industrial levels, it is obvious that an energy transition has to be realized not only in electricity-, heat-, and mobility sector, but in the industrial sector also. With 223 Mt CO₂-eq/a the European steel industry is responsible for around 5% of total greenhouse gas emissions (GHG) in the EU [1]. The large scale of the CO₂ emissions is due to the dependency on carbon based fossil energy sources for the conversion of iron ore into hot metal. Though there have been efforts to reduce the consumption of coke by producing steel over electric arc furnaces (EAF) mainly based on steel scrap - which amounts to 39% on the European steel production [2] - still 61% of the European steel production is still based on the CO₂-intensive blast furnace (BF)/converter route. Additionally to the named BF/converter production route, the combination of a direct reduction process (DRP) with an EAF represents a more environmental friendly process for steel production from iron ore. Currently, such processes are commercially available, but the market share is small due to the switch from the conventional BF/converter route to the DRP/EAF route comprising an enormous economic effort. Considering that, a gradual alteration of the process route should be taken into account. One attractive opportunity is the use of DRI to partly substitute iron ore loaded into a BF and consequently increase the metallization degree of the burden.

This way the DRP-inherent advantage of reduced CO₂ emissions can be utilized in a gradual manner, which lowers the economic commitment while keeping the plant operational. Furthermore, H₂ integration into the steel production processes in order to substitute fossil fuels is possible, which additionally reduces CO₂ emissions.

DRP's can be classified into gas-based and coal-based processes. Within the gas-based processes the most often represented are the Midrex and the HYL/Energiron technology [3].

Both processes take place in shaft furnaces operated optionally with natural gas, coke oven gas or syngas. In view of the aim to reduce GHG emissions, the HYL/Energiron process provides several advantages. Due to the in-situ reforming inside the shaft, the Energiron process contains no external catalytical reformer. Reforming, reduction and carburization proceed simultaneously which increases the energy efficiency compared to other direct reduction technologies due to an efficient heat integration. Furthermore, a larger fraction of the carbon input leaves the system via DRI in the form of Fe_3C , which results in a much lower reactivity of the DRI and enables a direct feed of freshly reduced hot DRI into the EAF. In addition to that, it reduces or eliminates the need of carbon injection to EAF. A further feature is the incorporation of a CO_2 and H_2O removal system as integral part of the reduction circuit. Benefits are an increased reduction potential of the recycled gas and less effort to use the occurring CO_2 stream in further processes. In consequence of the given characteristics, the present work is restricted to the HYL/Energiron process.

Related to the use of DRI in BFs various studies have been conducted. Tests with up to 90% DRI as iron-bearing charge were carried out [3]. The results show, that the use of pre-reduced burden materials has a positive influence on fuel consumption and the productivity of the BF. However, the use of DRI gradually loses its positive effects with increasing DRI charge what results in a decreasing CO_2 mitigation potential beginning of around 40% metallization of the iron loading of the blast furnace [4]. The possibility of using H_2 as a reducing agent to produce DRI of the time of this article has only rarely been reported [5]. The most comprehensive study by Yilmaz et al. [6] compared different types of DRI, including DRI partially produced by using hydrogen as reducing gas and their potential on the reduction of GHG emissions in a BF process. However, no modeling of the DRP was consulted. For the coupling of actual process models of a DRP and a BF, which is content of the present study, no reports could be found in the literature.

Process and Model development

Process scheme

Generally, a DRP/BF route with the integration of hydrogen can be divided into three main process steps: Hydrogen production, DRI production and hot metal production. A simplified process scheme is depicted in Figure 1.

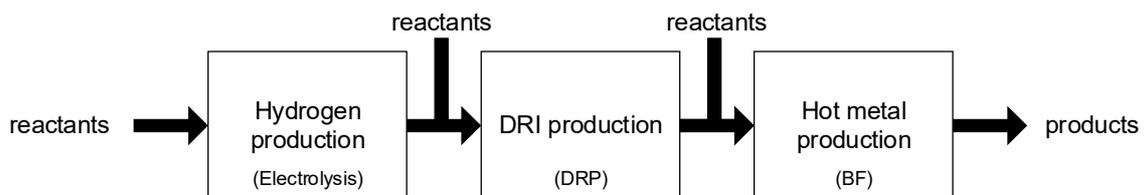


Figure 1 Simplified process scheme.

For the purpose of reducing CO_2 emissions, the production of hydrogen should be realized on the basis of water electrolysis. While the use of the current electricity mix is also considered in the present work, renewably generated electricity should be used in order to further the goal of this study, which is lessening the amount of CO_2 emissions. As mentioned before, for the production of DRI the HYL/Energiron process is applied. To complete the process chain the hot metal is produced in a BF.

By using DRI to partly supply the iron-bearing charge of the BF, two unique opportunities are offered. Besides the decrease of CO_2 emissions caused by a lower fuel consumption and the high amount of hydrogen compounds in natural gas, an increase in productivity of the BF can be achieved. For the present work, a steady-state model of the process chain was developed and applied, which allows for the estimation of fuel consumption and CO_2 emissions, though no kinetic process variables such as the productivity (t_{HM}/h) can be determined.

Modeling

The process was modeled using the flowsheet based simulation package Aspen Plus. The flowsheet was structured in a modular fashion that allows for the convenient substitution of

the process steps while ensuring constant boundary conditions. Advantage of the modular fashion is a simple replacement of single process steps for further studies, without developing an entirely new model.

Since the process mainly consists of process steps with solid phases, the property method SOLIDS was chosen for all blocks. Properties of the gaseous phase are calculated by Raoult's law, ideal gas law and Henry's law. The component data are derived from the Aspen native SOLIDS, INORGANIC and PURE36 databases.

Hydrogen production

The hydrogen used to substitute natural gas in the direct reduction plant as an example is supplied by a solid oxide electrolyser cell (SOEC). In order to drive the reactions an overall energy demand $\Delta_R h$ has to be met. In case of a reversible process, this energy demand consists of a thermal energy demand $T\Delta s$ and an electric energy demand $\Delta_R g$. In this work the SOEC is considered to be operating under thermoneutral conditions, which means that the electrical power supplied to the SOEC is used solely for the electrolysis reaction:

$$P_{el} = \Delta \dot{H}. \quad (1)$$

The SOEC model is based on the flowsheet introduced by Cinti et al. [7]. A scheme of the modeling approach is depicted in Figure 2.

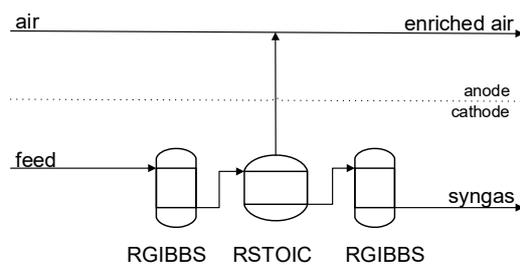


Figure 2 SOEC model according to Cinti et al. [7].

On the cathode side, the feed stream is first brought into chemical equilibrium in an RGIBBS block. Subsequently, water is converted to hydrogen in an RSTOIC block. Produced oxygen is separated from the product stream and routed to the anode side from where it is purged by an air stream. The second equilibrium block on the cathode side represents the electrodes catalytic activity.

DRI production

For the production of DRI, in the present work two different variations of an HYL/Energiron process are analyzed. A first version includes a gas-based preheating of the reducing gases corresponding to Figure 3a, which represents the current state of the art DRP. In a second version, an electrical preheater was applied corresponding to Figure 3b. For an HYL/Energiron process, the literature offers no suitable model, therefore process models for both variations were developed.

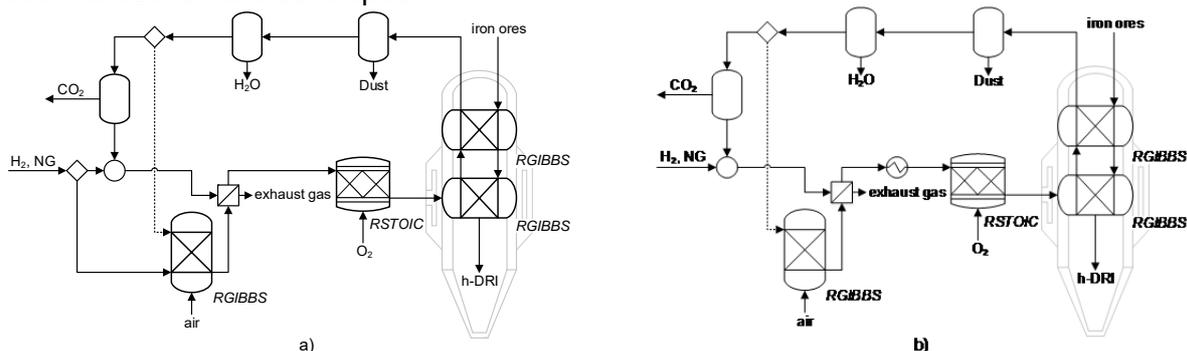


Figure 3 Simplified flowsheet of the DRP model with a) gas-heated and b) electrically heated reducing gases.

For both variations, the produced hydrogen is fed together with natural gas (NG) into the reduction-gas cycle where the gas mixture at first is preheated via a heat exchanger using the heat of the combustion of the purge-stream and an electrical or gas heater. Before the gas mixture enters the reactor shaft, a part of the NG is partially oxidized in an RSTOIC block to reach the necessary process temperature. The reactor shaft itself consists of two RGIBBS blocks. In the lower equilibrium block in which the reduction gas enters initially, mainly the carburization of the freshly reduced iron is realized. The reduction of iron ores mostly occurs in the upper equilibrium block. From off-gas leaving the shaft, dust and oxidants (H_2O , CO_2) are separated to increase the reduction potential. After a marginal part of the recycled gas is purged, the gas is mixed with freshly fed natural gas and optionally hydrogen.

To ensure that only realistic operating points are considered, the feed of NG was adjusted to maintain the thermal balance of the shaft. Therefore, constant heating losses of the shaft were assumed. To guarantee a constant product quality, the O_2 feed was adjusted to keep a constant amount of carbon inside the DRI. For the adjustment of the natural gas and oxygen stream, Aspen offers the native tool “design specifications”. Design specifications allow to tend a target variable to a target value depended on a varying value. To consider kinetic limitations of the iron oxide reduction inside an equilibrium block, a constant amount of wüstite was assumed to behave inert inside the reactor. Under this assumption, a constant metallization of the DRI was ensured.

The validity of the DRP model was verified with reference data based on actual operational data for the case of using natural gas solely as well as a gas-fed preheater. The input conditions for the reference data is listed in Table 1 and obtained from Ref. [8]. Table 2 shows simulation results compared to reference data. All values are related to the production of 1 ton of DRI. Compared to the reference data, the deviation of the simulation results was within 5%.

Table 1 Input values for the calculation.

Parameter	Value
Iron ore	1.35 t
Natural gas	9.96 GJ
O_2	60 Nm ³

Table 2 Comparison of simulation results with reference data.

Parameter	This work	Reference
M_{DRI}	94 %	94 % [9]
$C_{DRI, total}$	3.51 %	3.5 % [9]
$m_{CO_2, konz}$	256 kg	256 kg [9]
$m_{CO_2, em}$	165 kg	159 kg [8]
T_{DRI}	680 °C	675 °C [10]
T_{Topgas}	410 °C	407 °C [11]

Hot metal production

The model of a blast furnace that is used in the present work is derived from different models introduced by Yilmaz et al. [6] and Suzuki et al. [12]. A simplified flowsheet of the resulting model is depicted in Figure 4.

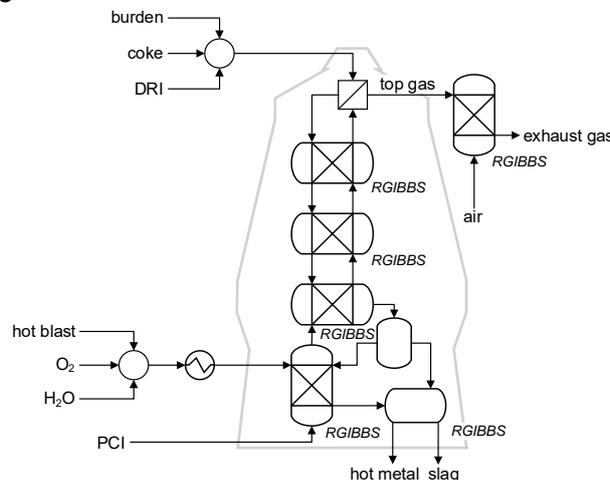


Figure 4 Simplified flowsheet of the blast furnace model.

In common blast furnace processes, solid materials are introduced from the top of the furnace whereas gaseous materials are fed at the lower part of the furnace together with pulverized coal (PCI). Sieverdingbeck et al. [13] developed a three-zone model using Aspen Plus, but also more current models for instance by Suzuki et al. [12] relate to this approach. Derived from the named literature models the present model of the BF is divided into three main reaction zones as well. Several other BF models have been based on this approach. Therefore, from top to bottom the furnace is separated into isothermal low-, middle-, and high-temperature equilibrium reactors. To consider the partial combustion of the PCI and a part of the descend coke load in near of the tuyere, another RGIBBS block is applied. To consider the later energetic use of the top gas, the included CO and H₂ are fully oxidized in another RGIBBS block. In the present work, it was assumed that the heat demand of the cowper is provided only by the top gas. Furthermore, the remaining heat was converted into electricity with an efficiency of $\eta_{PP} = 0.4$ to serve the power demand of the compressors and the air separation unit.

Analogous to the model of the DRP, to guarantee a constant product quality as well as considering only realistic operating points, several design specifications are necessary. Firstly, the feed of iron oxides into the BF was adjusted to keep the amount of hot metal constant when feeding DRI. To maintain the thermal balance of the BF, the feed of the hot blast was adjusted. To guarantee a constant amount of C inside the hot metal, the coke rate was adjusted while the amount of PCI was kept at a constant level. Furthermore, the O₂ feed was adapted to keep a constant adiabatic flame temperature of 2150 °C.

Process

For the operation of both process models, several estimates for the process control have to be made. Gaseous inlet streams of the model of the DRP as well as the model of the BF have to be compressed to certain pressure levels according to Table 3 and Table 4. All compressors were modeled with an isentropic efficiency of $\eta_{isentropic} = 0.72$. Both models comprise the simplification, that the pressure drop in the various process steps is not considered. In the case of a gas-heated reduction stream, the efficiency of the heater is set to $\eta = 0.75$. The electrical heater was assumed to be ideal.

For the heat exchanger in the preheating zone of the blast furnace, a constant temperature difference between the hot outlet and cold inlet derived from the basic case of the blast furnace was assumed. An overview over the operating conditions of the main process steps is given in Table 3 and Table 4.

Table 3 Operating conditions of the DRP model.

Boundary condition	Value	Description
p_{DRP}	7 bar	Pressure level
$\omega_{Fe, Pellets}$	67.3 %	Iron amount in pellets

Table 4 Operating conditions of the BF model.

Boundary condition	Value	Description
ϑ_{HB}	1250 °C	Hot blast temperature
ϑ_{ad}	2150 °C	Adiabatic flame temperature
p_{HB}	4.2 bar	Hot blast pressure level
p_{TG}	2.5 bar	Top gas pressure level
$\omega_{Fe, Iron Ore}$	59.2 %	Iron amount in pellets

The DRP model as well as the blast furnace model contain several recycles. These as well as the above mentioned design specifications cause calculation loops. In order to achieve reliable results, design specifications were always nested outside, tear streams inside. The tolerance for the outermost calculation loop was set to 10^{-5} . With every further level of nesting, the tolerance was decreased by one order of magnitude. Simulations were carried out with different initial values to exclude the occurrence of local minima.

Results and Discussion

Maximum H₂ input

For a qualitative assessment of the potential to reduce CO₂ emissions via the BF route by the integration of H₂, a characteristic value is the boundary percentage of H₂ that can be fed into the process without negative impact on DRI quality. As depicted in Figure 5, up to a volumetric fraction of H₂ of approximately $\phi_{H_2} = 0.65$ which equals $\chi_{H_2} = 0.36$ of the chemical input power of the DRP, the carbon content in the DRI is unaffected. With a further increase in H₂, the carbon content declines to $\omega_C = 0$ at around $\phi_{H_2} = 0.85$. Because of the decreasing carbon content in DRI, the fuel rate of the BF has to be increased to reduce the remaining oxides inside the DRI. Therefore, in the present work the simulated range was kept below $\phi_{H_2} = 0.65$.

The decrease of the carbon content between $\phi_{H_2} = 0.65$ and $\phi_{H_2} = 0.85$ seems comparatively rapid. For further investigations, in case of evaluating also volumetric fractions of more than $\phi_{H_2} = 0.65$, the use of the software ChemApp using FactSage methods and physical property data for the realization of equilibrium calculations will take place.

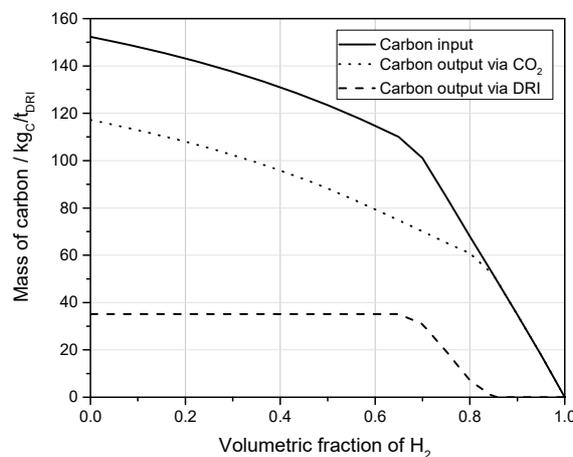


Figure 5 Input and output mass of carbon into the DRP per ton of DRI.

CO₂ emission reduction

For the evaluation of the CO₂ emissions of the process chain a base case was used, which is considering hot metal production only via BF. The CO₂ emissions of the base case were calculated to be $m_{CO_2} = 1365 \text{ kg}_{CO_2}/t_{HM}$, compared to a literature value of $m_{CO_2} = 1400 \text{ kg}_{CO_2}/t_{HM}$ [12]. As mentioned before, the top gas was burned completely to obtain the total CO₂ emissions of the process. Based on the assumption that the heat demand of the cowper as well as the power demand of the compressors and the air separation unit of the BF are fully covered by the energy-related use of the top gas, there are no CO₂ emissions considered for these process units. For the power supply, two different cases were examined. The first case is characterized by the use of eco power. In this case CO₂ emissions were assumed to be zero. In the second case, a typical power mix was assumed. The CO₂ emission factor was set to $0.534 \text{ kg}_{CO_2}/\text{kWh}_{el}$ according to the Federal Environmental Agency of Germany (UBA) [14]. To consider emissions of the air separation unit for the DRP in case of using power mix, a factor of $0.36 \text{ kWh}_{el}/\text{Nm}^3_{O_2}$ according to Ref. [15] was assumed. As mentioned before, the CO₂ mitigation potential decreases starting at 40% metallization, wherefore for the present paper the simulation range was set between $m_{DRI} = 0 \text{ kg}_{DRI}/t_{HM}$ and $m_{DRI} = 400 \text{ kg}_{DRI}/t_{HM}$.

DRI production via natural gas

In a first case, the DRP was operated only with natural gas. The preheating of reducing gases was assumed to be gas-based. As depicted in Figure 6, a linear decrease of CO₂ emissions with a gradient of approximately $m_{CO_2} = 55 \text{ kg}_{CO_2}$ per $m_{DRI} = 100 \text{ kg}_{DRI}$ that are fed to the BF takes place. This leads to a maximum CO₂ emission mitigation of 220 kg_{CO_2} per ton of hot metal within the simulation area, which is equivalent to a CO₂ emission reduction of 16.1 % compared to the base case.

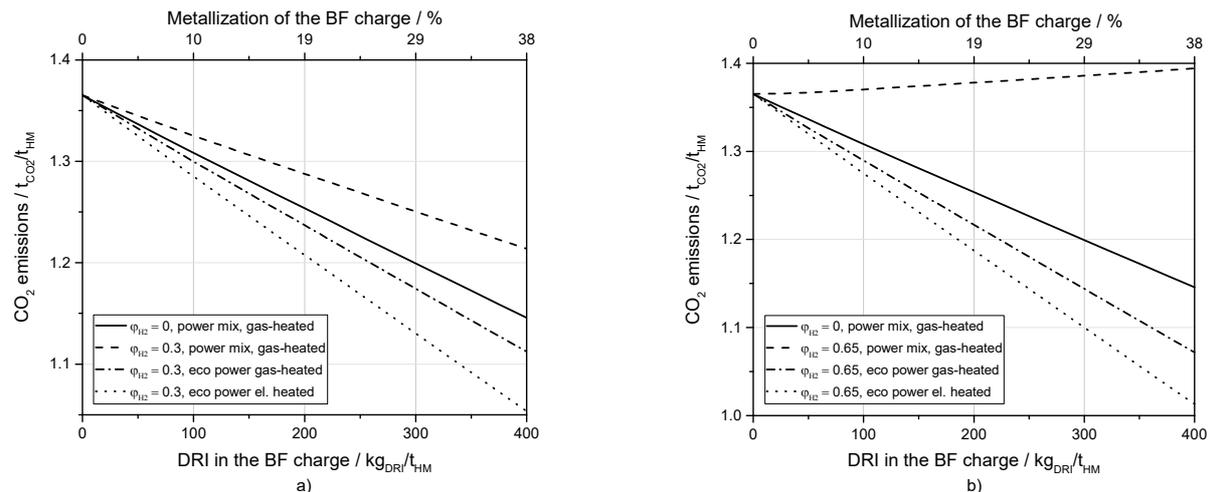


Figure 6 CO₂ emissions for different plant variations using a) $\varphi_{H_2} = 0$ and b) $\varphi_{H_2} = 0.65$.

Hydrogen production via power mix

For the second case hydrogen was used to substitute a part of natural gas and was generated using typical power mix. The gas-based preheater was maintained. Figure 6a depicts a substitution of natural gas with hydrogen of $\varphi_{H_2} = 0.3$ ($\chi = 0.115$), while in Figure 6b the amount of hydrogen was set to $\varphi_{H_2} = 0.65$ ($\chi = 0.36$). As can be seen based on Figure 6 a), for $\varphi_{H_2} = 0.3$ the CO₂ emission reduction potential lowers compared to the case of DRI production only via natural gas. For $\varphi_{H_2} = 0.65$ CO₂ emissions even increase. For a substitution of $\varphi_{H_2} = 0.3$ a linear decrease of CO₂ emissions with $m_{CO_2} = 37.9 \text{ kg}_{CO_2}$ per $m_{DRI} = 100 \text{ kg}_{DRI}$ takes place. This leads to a maximum CO₂ emission mitigation of $m_{CO_2} = 151.4 \text{ kg}$ per ton of hot metal, what results in a CO₂ emission reduction of 11.1% compared to the base case in the simulated range. A substitution of $\varphi_{H_2} = 0.65$ leads to an increase of $m_{CO_2} = 7.3 \text{ kg}_{CO_2}$ per $m_{DRI} = 100 \text{ kg}_{DRI}$. It is shown, that the use of hydrogen as a reducing agent in the DRP under the use of a typical power mix does not entail a further reduction of CO₂ emissions.

Hydrogen production via eco power

In a third case the production of hydrogen was estimated to be realized using eco power, while the preheating still was estimated as gas-based. As depicted in Figure 6 a) and b), the integration of hydrogen into the process under the use of eco power allows for a further reduction of CO₂ emissions compared to the case of DRI production only via natural gas. For a substitution of $\varphi_{H_2} = 0.3$ a linear decrease of CO₂ emissions with $m_{CO_2} = 63.2 \text{ kg}_{CO_2}$ per $m_{DRI} = 100 \text{ kg}_{DRI}$ takes place. Due to an increased substitution of $\varphi_{H_2} = 0.65$ the CO₂ emission reduction potential increases to $m_{CO_2} = 73.3 \text{ kg}_{CO_2}$ per $m_{DRI} = 100 \text{ kg}_{DRI}$. As a result a maximum CO₂ emission mitigation of $m_{CO_2} = 252.9 \text{ kg}$ respectively $m_{CO_2} = 293.3 \text{ kg}$ CO₂ per ton of hot metal can be achieved, which is equivalent to a CO₂ emission reduction of 18.5 % and 21.5 % respectively compared to the base case.

Use of an electrical preheater

The fourth case was conducted under the estimation of the use of eco power for the production of hydrogen as well as an electrical preheater replacing the gas-based preheater. For $\varphi_{H_2} = 0.3$ of hydrogen as well as for $\varphi_{H_2} = 0.65$ the reduction potential for CO₂ emissions further increases compared to the case of using a gas-based preheater. By comparing Figure 6 a) and b) it is also obvious that the amount of the CO₂ emission reduction potential, which is caused by the use of hydrogen as a reducing agent increases with increasing amounts of hydrogen compared to the impact of the type of the preheater. For a substitution of $\varphi_{H_2} = 0.3$, a linear decrease of CO₂ emissions with $m_{CO_2} = 77.9 \text{ kg}_{CO_2}$ per $m_{DRI} = 100 \text{ kg}_{DRI}$ takes place. A substitution of $\varphi_{H_2} = 0.65$ further increases CO₂ emission reduction potential to 88 kg_{CO_2} per 100 kg_{DRI} . This leads to a maximum CO₂ emission mitigation of 311.6 kg and 352 kg CO₂ per ton of hot metal respectively, what results in a CO₂ emission reduction of 22.8 % and

25.8 % respectively compared to the base case.

Impact on energy input and output

Beside the CO₂ reduction potential of the different cases it is also of interest how the energy balance of the system changes for the use of DRI as a pre-reduced input material for the BF compared to the base case. Figure 7 depicts the relevant energy input and output for the DRP/BF route.

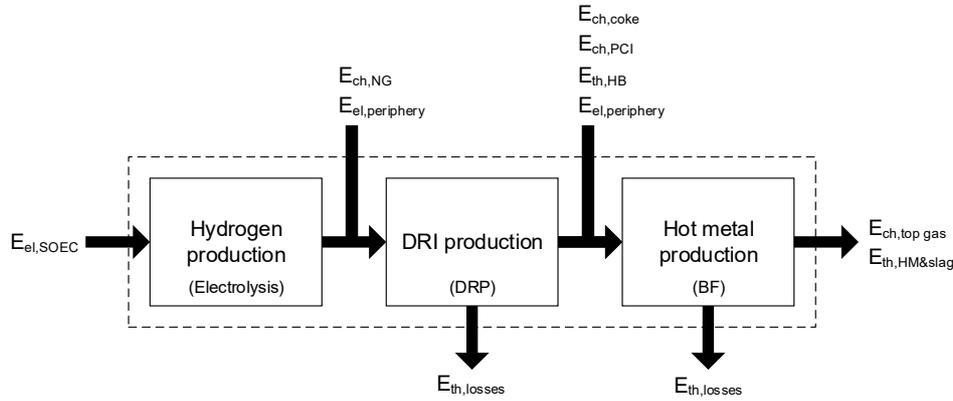


Figure 7 Energy input and output for the DRP/BF route.

Figure 8 a) and b) show the specific energy input and output for the base case compared to the specific energy input and output for different plant variations using 400kg DRI per ton of hot metal. For case A, the production of DRI only via natural gas and a gas-based preheater was estimated. Case B represents the case of substituting natural gas by hydrogen to $\varphi_{H_2} = 0.3$ and variation C a substitution of $\varphi_{H_2} = 0.65$. In case of using hydrogen, an electric preheater as well as the use of eco power were estimated. As shown in Figure 8, when using DRI, the specific energy input per ton of hot metal decreases slightly relative to the base case.

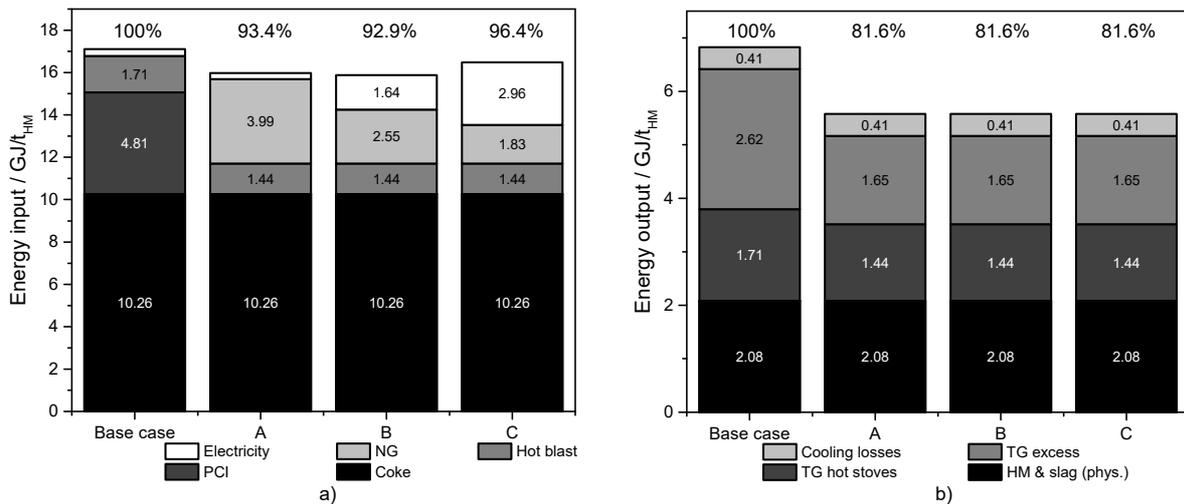


Figure 8 Specific energy input (a) and output (b) for the use of 400kg_{DRI}/t_{HM}.

For variation B and C the chemical energy input to the DRP consisting of natural gas and hydrogen compared to variation A slightly decreases, but due to the efficiency of the electrolysis and an increasing demand of heat for the electrical preheating of reducing gases, for variation B the savings are negated and for variation C the demand even increases. However, even for variation C the energy input is smaller than in the base case, but it has to be considered, that also the specific energy output per ton of produced hot metal decreases, since composition and mass of the generated top gas changes when using DRI in the BF.

Conclusion

In this paper, the CO₂ emissions of different plant variations for the DRP/BF route were compared. Furthermore, the impact of using DRI as pre-reduced input to the BF on the specific energy input was investigated. The results showed that the use of DRI to pre-reduce parts of the BF burden from a technical point of view is a practical way to realize a stepwise change from the BF/converter-route to a DRP/EAF route. For the use of hydrogen it could be demonstrated, that substitution of parts of the natural gas feed to the DRP is not inherently a useful opportunity to mitigate CO₂ emissions. The central aspect is the use of emission free eco power for the production of hydrogen. In addition, the type of the preheater was identified to be of significantly influence for CO₂ emission mitigation. Thereby, a further CO₂ emission mitigation potential by using an electrical heater as well as eco power could be shown. Within the simulated range, a maximum CO₂ emission mitigation potential of 25.8% relative to the base case was found.

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