**ORIGINAL ARTICLE** 



# Evaluation of biomass-based production of below zero emission reducing gas for the iron and steel industry

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### Abstract

The present paper focuses on the production of a below zero emission reducing gas for use in raw iron production. The biomassbased concept of sorption-enhanced reforming combined with oxyfuel combustion constitutes an additional opportunity for selective separation of  $CO_2$ . First experimental results from the test plant at TU Wien (100 kW) have been implemented. Based on these results, it could be demonstrated that the biomass-based product gas fulfills all requirements for the use in direct reduction plants and a concept for the commercial-scale use was developed. Additionally, the profitability of the below zero emission reducing gas concept within a techno-economic assessment is investigated. The results of the techno-economic assessment show that the production of biomass-based reducing gas can compete with the conventional natural gas route, if the required oxygen is delivered by an existing air separation unit and the utilization of the separated  $CO_2$  is possible. The production costs of the biomass-based reducing gas are in the range of natural gas-based reducing gas and twice as high as the production of fossil coke in a coke oven plant. The  $CO_2$  footprint of a direct reduction plant fed with biomass-based reducing gas is more than 80% lower compared with the conventional blast furnace route and could be even more if carbon capture and utilization is applied. Therefore, the biomass-based production of reducing gas could definitely make a reasonable contribution to a reduction of fossil  $CO_2$  emissions within the iron and steel sector in Austria.

Keywords Iron and steel  $\cdot$  Low-carbon steelmaking  $\cdot$  Direct reduction  $\cdot$  Biomass  $\cdot$  Sorption-enhanced reforming  $\cdot$  Oxyfuel combustion

# 1 Introduction

Today the iron and steel industry in EU-28 is responsible for 200 million tons of carbon dioxide [1] which amounts to a share of 5% of the total carbon dioxide equivalent ( $CO_2e$ ) [2] emissions [3]. These numbers show that especially the transformation of heavy load industries like the iron and steel industry towards low-carbon technologies will be challenging. In Austria the iron and steel industry also contributes to a significant share concerning greenhouse gas emissions. In 2017, 8.1 million tons of crude steel were produced in Austria [4], which are responsible for around 16% of the total greenhouse gas emissions [5]. Technological development

has enabled to improve the energy efficiency and to reduce  $CO_2$  emissions in this sector. However, the principles of steelmaking have not changed fundamentally over the years. In 2017, over 91% of the Austrian crude steel was produced within oxygen-blown converters, which were fed with hot metal from blast furnaces. The remaining share was produced within electric arc furnaces [4]. According to the EU Roadmap 2050 [6], the  $CO_2$  emissions within the iron and steel industry must be reduced by around 85%. To accomplish this major goal, a complete conversion towards low-carbon steelmaking technologies has to be done.

Numerous researchers and international institutions investigate alternative low-carbon steelmaking routes. Especially, the ULCOS program [7, 8] has evaluated the  $CO_2$  reduction potential of over 80 existing and potential technologies. Several investigations are working on further optimization of fossil fuel-based state-of-the-art processes like the coke and pulverized coal-based-integrated blast furnace route [9–11]. All this optimization steps to reduce the consumption of fossil fuels are limited [12]. For reaching the previous described

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climate goals within the iron and steel sector, a fundamental change of steelmaking is necessary. The ULCOS program [7, 8] identified four technologies with CO<sub>2</sub> emission reduction potentials of more than 50%. The technologies within this program, which are based on carbon capture and storage (CCS) or utilization (CCU), are the top-gas recycling within the blast furnace (BF-TGR-CCS/U), a novel bath-smelting technology (HISARNA-CCS/U) [13, 14], and a novel direct reduction process (ULCORED-CCS/U). Only the novel ULCOLYSIS [15] process, which is characterized by melting iron ore through electric direct reduction, is not based on CCS or CCU. In addition to the research activities in Europe, the COURSE50 program in Japan, POSCO in Korea, AISI in the USA, and the Australian program are some international examples for investigations regarding CO<sub>2</sub> reduction in the iron and steel industry [16]. The COURSE50 program [8, 16, 17] is focused on H<sub>2</sub>-based reducing agents in blast furnace (BF) for decreasing the fossil coke consumption and technologies for capturing, separating, and recovering CO<sub>2</sub> from the BF gas. POSCO [8, 16, 18] in Korea is working on the adaptation of CCS and CCU to smelting reduction processes, like the FINEX and COREX process. Furthermore, POSCO is researching in bio-slag utilization, pre-reduction and heat recovery of hot sinter, CO<sub>2</sub> absorption using ammonia scrubber, hydrogen production out of coke-oven gas (COG), and iron ore reduction using hydrogen-enriched syngas. AISI [8, 16] is working on the molten oxide electrolysis, which is similar to the ULCOLYSIS concept and iron making by hydrogen flash smelting. The research programs regarding breakthrough iron and steelmaking technologies in Brazil, Canada, and Australia [19] are all strongly focused on biomass-based iron and steel production routes for replacing fossil coal and coke by use of biomass-derived chars as substitutes [8, 16, 20].

Summing up, there are a lot of investigations going on around the world to reduce the  $CO_2$  footprint of the iron and steel industry.

The most of the previous described concepts apply CCS or CCU to reach a  $CO_2$  reduction potential over 50% in comparison to the conventional integrated BF route. Nevertheless, the implementation of CCS requires a fundamental investigation due to storage sites and long-term response of the environment. Beside the CCS or CCU-based approaches, the replacement of fossil fuel-based reducing agents by biomass-based substitutes or the use of hydrogen as reducing agent are promising approaches for reaching the climate targets within the iron and steel sector. Furthermore, some electric direct reduction processes like ULCOWIN, MOE, and ULCOLYSIS are under investigation. One possible  $CO_2$  reduction path could also be the rise of the share of steel production through electric arc furnaces. Therefore, enough high-quality scrap must be available.

With respect to the estimates regarding biomass potential in the next decades [20, 21], in Austria beside the rise of the share of steel production through scrap-based electric arc furnaces, another possible synergetic transition option seems to be the replacement of the integrated blast furnace route with the direct reduction of iron ore based on biomass-based reducing gas. The Austrian steel manufacturing and processing group, voestalpine AG, is already operating one of the biggest direct reduction plants, based on the MIDREX concept and reformed natural gas as reducing agent in Texas [22]. This approach would combine the gained expertise within the field of direct reduction with the Austriadeveloped concept of dual fluidized bed steam gasification [23]. Within the present work, a biomass-based production of biogenic reducing gas through dual fluidized bed steam gasification, which allows the replacement of steam reformed natural gas, is investigated. At this stage, it remains unclear if the investigated process is competitive with respect to other production routes for the supply of reducing gas for iron ore reduction.

So far, following question has not been answered sufficiently:

How can the production of biomass-based reducing gas via dual fluidized bed steam gasification enable a reasonable contribution to a reduction of fossil  $CO_2$  emissions within the iron and steel sector?

The following paper describes the results of the investigated process enabling the production of a below zero emission reducing gas by applying the biomass-based dual fluidized bed steam gasification technology in combination with carbon capture and utilization. The investigations are based on experimental results combined with simulation work. The present paper discusses:

- The comparison of different iron- and steelmaking routes regarding their CO<sub>2</sub> footprint
- The *proposed process concept* for the production of biomass-based reducing gas
- Experimental and simulation results achieved
- The results of a techno-economic assessment

# 2 Concept and methodology

With regard to the techno-economic assessment of the selective separation of  $CO_2$  technology OxySER, a plant concept for the integration in a direct reduction process has been developed. Beforehand, a short overview and comparison of primary and secondary iron and steelmaking routes regarding their  $CO_2$  footprints will be given. Furthermore, the application of dual fluidized bed steam gasification with respect to the combination of sorption-enhanced reforming and oxyfuel combustion will be explained.

# 2.1 Comparison of iron and steelmaking routes regarding their CO<sub>2</sub> footprint

Two main steelmaking processes can be distinguished. The primary steelmaking route converts virgin iron ores into crude steel (CS). Secondary steelmaking is characterized by the recycling of iron and steel scrap in an electric arc furnace [8, 24]. Table 1 gives an overview of chosen iron and steelmaking routes and the comparison regarding CO<sub>2</sub> footprint. First of all, the primary steelmaking integrated blast furnace (BF) route, which is predominant in Austria. Thereby, steel production takes place at an integrated steel plant, where iron ores are reduced into hot metal through the use of reduction agents such as coke or coal. Afterwards, the hot metal is converted into steel by oxygen injection in a basic oxygen furnace (BOF). As result of the high energy demand of 11.4  $GJ/t_{CS}$ on fossil reducing agents, the CO<sub>2</sub> footprint of the BF-BOF route is with 1.694 t  $CO_2e/t_{CS}$  very high [25]. Furthermore, the secondary steelmaking electric arc furnace (EAF) route is used in Austria. Therein, the major feedstock is ferrous scrap, which is melted mainly through the use of electricity. However, increasing the share of EAF steel is constrained by the availability of scrap, and the quality requirements for steel grades have to meet [8]. The smelting reduction route belongs also to the state-of-the-art iron and steelmaking routes. Within this route, iron ores are heated and prereduced by the off-gas coming from the smelter-gasifier. The pre-reduction step could be realized in a shaft kiln (COREX) or a fluidized bed reactor (FINEX). Pre-reduced iron ores are then melted in the smelter-gasifier. The smelter-gasifier uses oxygen and coal as a reducing agent. Afterwards, the hot metal is also fed to the BOF for steelmaking. Another possibility of steelmaking is the primary direct reduction (DR) route. MIDREX is one of the used direct reduction technologies. It is characterized by the reduction of iron ores into solid direct reduced iron (DRI) within a shaft kiln. The direct reduction technologies could also work within a fluidized bed reactor. Examples include the FINMET and CIRORED process [38]. The direct reduction is driven by the fed of a reducing gas. Currently, the commercial used reducing gas is based on the reforming of natural gas. For extended information regarding the fundamentals of iron and steelmaking routes, a reference is made to [8, 24, 39].

Beside the previous described state-of-the-art iron and steelmaking routes, some innovative developments and investigations are compared with the conventional routes regarding their energy demand,  $CO_2$  footprint, merit, and demerit in Table 1. Therein, the integrated blast furnace route (BF and BOF) which is predominant in Austria is set as reference regarding  $CO_2$  emissions. Recycling of the blast furnace top-gas in combination with CCS or CCU (BF-TGR-CCS/U and BOF) or the replacement of fossil coal by biogenic substitutes reduces the fossil reducing agent demand and decrease the  $CO_2$  footprint of integrated blast furnace routes up to 50% [7, 16, 26, 30, 31].

The replacement of the BF by smelting reduction processes like the COREX or FINEX process would raise slightly the CO<sub>2</sub> footprint due to the high consumption of fossil coal. An ecologically favorable operation of smelting reduction processes only could be realized by the use of CCS or CCU [8, 16, 18]. The use of a smelting reduction technology based on bath-smelting (HISARNA-CCS/U and EAF) in combination with CCS would reduce the CO<sub>2</sub> emissions up to 80% [7, 16].

Direct reduction plants enable a big CO<sub>2</sub> emission saving potential in comparison with the integrated BF route due to the present used reformed natural gas as reducing agent. Reformed natural gas consists to a large extent of hydrogen, which results in lower CO<sub>2</sub> emissions due to the oxidation of hydrogen to steam within the reduction process [12]. The replacement of the integrated BF route by the state-of-theart MIDREX plant, which is based on the reduction of iron ore within a shaft kiln by the use of reformed natural gas, would decrease the CO<sub>2</sub> emissions by 50% in comparison with the reference route [12, 32, 33]. The economic viability of direct reduction-based routes, which are based on reformed natural gas, strongly depend on the natural gas price which is in Europe much higher than in North America [33]. Within the ULCOS project, a novel direct reduction process (ULCORED-CCS/U) based on partial oxidized natural gas is investigated [7, 8]. By the reduction of the required amount of natural gas and the application of CCS or CCU, the CO<sub>2</sub> emissions could be decreased up to 65% compared with the reference route. The dual fluidized bed steam gasification process, based on the bed material limestone, which is called sorption-enhanced reforming (SER), produces a biomassbased hydrogen-rich gas, which allows the replacement of the steam reforming unit for reforming of natural gas. The application of SER to produce a biomass-based reducing gas for the MIDREX process (MIDREX-BG-SER) reduces the CO<sub>2</sub> footprint compared with the integrated BF route up to 80%. The combination of SER with oxyfuel combustion (OxySER) enables an in situ CO<sub>2</sub> sorption within the reducing gas production process. Beside the production of biomass-based reducing gas, a CCU or CCS ready CO<sub>2</sub> stream is released. Therefore, a below zero emission reducing gas due to the application of CCU or CCS is generated. Another direct reduction breakthrough technology could be the HYBRIT process, which is based on the reducing agent hydrogen, produced by electrolysis [16, 26, 34, 35]. Therefore, the emissions within the HYBRIT process are mostly caused by the CO<sub>2</sub> footprint of the electricity mix. With regard to the Austrian electricity mix, with a CO<sub>2</sub> footprint of 0.218 kg CO<sub>2</sub>e/kWh<sub>el</sub> [36], a CO<sub>2</sub> emission saving potential up to 50% could be reached with the HYBRIT process.

Table 1 Overview of different	Overview of different iron and steelmaking routes including their energy demands and CO2 emissions [16]	including their energy de	emands an	d CO <sub>2</sub> emis	ssions [16]					
Iron and steelmaking route <sup>1</sup>	Description of the technology	Reducing agent	Total energy demand [GJ/t <sub>CS</sub> ]	Energy demand reducing agent [GJ/t <sub>CS</sub> ]	N et power demand [GJ/t <sub>CS</sub> ]	CO <sub>2</sub> emissions <sup>2</sup> [t CO <sub>2</sub> e/ t <sub>CS</sub> ]	Savings potential <sup>2</sup> [%CO <sub>2</sub> e]	Advantages	Disadvantages	Literature
Integrated Blast Furnace route BF and BOF	Fossil reducing agents (coke and pulverized coal) are used as reducing agents; mixture of sinter, pellets, and additives are fed to the BF	Coke/coal	19.2	15.3	0.6	1.694	I	Production of HM in existing BF for steelmaking is the most cost-efficient technology to-	State of the art BFs are operated near their theoretically minimum energy limit, further CO <sub>2</sub> reductions are difficult	[25-27]
BF-TGR-CCS/U and BOF	Upgraded and recirculated BF gas is used as reducing agent, parts of pulverized coal and top-charged coke are replaced, CCS can be used for further CO <sub>2</sub> reduction	Coke/coal/recirculated BF gas	20.0	11.6	1.4	0.813	- 52%	day Top-gas recycling captures CO <sub>2</sub> and enables CCS, Reduction of fossil reducing agent demand	Higher total energy demand because of additional energy demand for carbon capture, higher net energy demand due to the lack of power recovery from BF	[7, 15, 26, 28, 29]
BF-Bio-Char and BOF	Charcoal replaces fossil coal by 100% in the BF	Coke/charcoal	19.2	15.3	0.6	1.220	- 28%	Replacement of fossil coal by charcoal could be quite straightforward	gas Charcoal is more expensive than fossil coal, handling, transportation and storage is more difficult compared with fossil coal	[16, 26, 30, 31]
Smelting reduction route COREX and BOF	Combination of pre-reduction in a shaft kiln and smelter gasifier	Coal	17.7	15.9	0.6	1.975	+17%	No need for coke oven plant	Restrictions for non-coking coal quality, customer for export gas necessary for economic viabil-	[12, 16, 26]
FINEX and BOF	Combination of pre-reduction in a flu- idized bed reactor and smelter-gasifier	coal	I	I	I	1.910	+ 13%	No need for coke oven plant, pelletizing, sintering or agglomeration of iron-bearing	ity Technology not wide-spread	[12, 16]
HISARNA-CCS/U and EAF	Bath-smelting technology which combines coal	Coal	18.0	15.0	2.5	0.330	-81%	materials No need for coke oven and	Technology at demonstration stage,	[7, 16]

Table 1 (continued)										
Iron and steelmaking route <sup>1</sup>	Description of the technology	Reducing agent	Total energy demand [GJ/t <sub>CS</sub> ]	Energy demand reducing agent [GJ/t <sub>CS</sub> ]	N et power demand [GJ/t <sub>CS</sub> ]	CO <sub>2</sub> emissions <sup>2</sup> [t CO <sub>2</sub> e/ t <sub>Cs</sub> ]	Savings potential <sup>2</sup> [%CO <sub>2</sub> e]	Advantages	Disadvantages	Literature
Direct reduction route	pre-heating and partial pyrolysis in a reactor, a smelter vessel is used for final ore reduction and a melting cyclone for ore smelting							sinter/pellet plant. Use of non-coking coal qualities. Economic via- ble even at small size	more net power demand because of EAF and CCS	
MIDREX and EAF	DRI production based on shaft furnace with reformed natural gas as reducing agent. Based on hum/neller ore	Reformed natural gas	16.6	10.0	2.8	0.835	-51%	No need for coke oven and sinter plant	OPEX strongly depend on natural gas price, which is very different around the worlf	[12, 32, 33]
ULCORED-CCS/U and EAF	DRI production based on shaft furnace with partial oxidized natural gas as reducing agent. In situ CO <sub>2</sub> removal system. Use of water gas shift reactor into high H <sub>2</sub> contents in the reducing shaft	Partial oxidized natural gas				0.600	- 65%	Reduction of natural gas consumption helps to reduce OPEX	Requires pure oxygen instead of air. Technology at demonstration stage	[7, 8]
MIDREX-BG-SER and EAF	Reformed natural gas is replaced by biomass-based reduc- ing gas produced by dual fluidized bed steam gasification	Biomass-based reducing gas	16.6	10.0	2.8	0.280*	- 83 %	Replacement of reformed natural gas by biomass-based reducing gas could be quite	OPEX strongly depend on biomass price	Captured within this paper
MIDREX-BG-OxySER-CCS/U and EAF	Reformed natural gas is replaced by biomass-based reduc- ing gas produced by dual fluidized bed steam gasification and in situ CO <sub>2</sub> capture and utilization	Biomass-based reducing gas		10.0		Below zero	More than 100% reduction	Replacement of reformed natural gas by biomass-based reducing gas could be quite straightforward	OPEX strongly depend on biomass price. CCS or CCU approach requires pure oxygen instead of air as fluidization agent in the	Captured within this paper
HYBRIT and EAF	Hydrogen produced with water electrolysis is used as reducing agent	Hydrogen	14.7	6.8	12.6	0.800	- 53%	CO <sub>2</sub> emissions very low if renewable energy sources	Hydrogen production is quite expensive with current technologies and	[16, 26, 34, 35]

Table 1 (continued)										
Iron and steelmaking route <sup>1</sup>	Description of the technology	Reducing agent	Total energy demand [GJ/t <sub>CS</sub> ]	Energy demand reducing agent [GJ/t <sub>CS</sub> ]	N et power demand [GJ/t <sub>CS</sub> ]	CO <sub>2</sub> emissions <sup>2</sup> [t CO <sub>2</sub> e/ t <sub>CS</sub> ]	Savings potential <sup>2</sup> [%CO <sub>2</sub> e]	Advantages	Disadvantages	Literature
Secondary steelmaking route	within a direct reduction plant							are used. Almost zero direct emissions from production	requires a lot of electrical energy. Technology at demonstration stage	
EAF	Instead of DRI, scrap is used as iron source	Electricity	3. 3		2.1	0.190	~89%	DRI production is replaced by recycling of ferrous scrap. Almost zero direct emissions from production	Increasing the share of [12, 26] EAF steel is constrained by the availability of high quality scrap	[12, 26]
Other steelmaking routes ULCOLYSIS	Melting iron ore at 1600 °C by using electric direct reduction	Electricity	15.0		13.0	0.800	- 53%	CO <sub>2</sub> emissions very low if renewable energy sources are used. Almost zero direct emissions from production	Technology at lab scale. High net power demand	[7, 16]
<sup>1</sup> Energy values and CO <sub>2</sub> emissions includes material preparation, ironmaking, steelmaking, and casting/based on 80% hot metal or DRI and 20% scrap except from secondary steelmaking route: 100% scrap in EAF <sup>2</sup> CO <sub>2</sub> emission factor for grid/calculation model: 0.218 kg CO <sub>2</sub> <i>e</i> /kWh <sub>el</sub> (Austrian electricity mix)/saving potential in comparison with BF and BOF route as reference [36]	ons includes material prepara alculation model: 0.218 kg C	tion, ironmaking, steelm. O <sub>2</sub> e/kWh <sub>el</sub> (Austrian ele	aking, and ctricity mix	casting/base )/saving po	ed on 80% tential in co	hot metal or omparison w	DRI and 20% (th BF and B	<ul> <li>scrap except from :</li> <li>DF route as reference</li> </ul>	secondary steelmaking ru e [36]	ute: 100%

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\* Assumptions: energy values are the same as within the MIDREX route, and the  $CO_2$  emissions are calculated by the difference between the emissions from the MIDREX route based on natural gas and the caused emissions only through natural gas as reducing gas [37]

\*\* Assumptions: energy values are the same as within the MIDREX route with CCS [7] and the CO<sub>2</sub> emissions are below zero because of the combination from the use of a biomass-based feedstock with CCS or CCU

Further possibilities are the rise of the share of steel production through scrap-based electric arc furnaces. This steelmaking route enables  $CO_2$  reduction potentials up to 90%, because of the replacement from ironmaking processes with scrap. The EAF-based routes are strongly depended on the availability of high-quality scrap [12, 26]. Furthermore, some novel electric direct reduction processes, like the ULCOLYSIS project, are under investigation [7, 16]. Similar to the HYBRIT process, the electric direct reduction processes are strongly depended on the  $CO_2$  footprint of the national electricity mix, because of the high-net power demands.

Several technologies provide the possibility of additional carbon-emission reduction by sequestration of  $CO_2$ . The use of post-combustion capture technologies, like pressure swing adsorption or amine scrubber, is the possibility for the sequestration of  $CO_2$  within iron and steelmaking routes [40]. Within the OxySER process, through the in situ  $CO_2$  sorption, a CCU or CCS ready  $CO_2$  stream is produced. Further explanations regarding  $CO_2$  sequestration can be found in [41–43]. The selective separated and purified  $CO_2$  could be used in further process steps as raw material, *carbon capture and utilization*, or stored in underground deposits, *carbon capture and storage* [43, 44].

Today around 230 million tons of carbon dioxide per year are globally utilized materially. One hundred thirty million tons are used in urea manufacturing and 80 million tons for enhanced oil recovery [45]. With the assumption that hydrogen for the ammoniac production is produced by water electrolysis, which is beside CO<sub>2</sub> the primary energy source for urea production, external CO<sub>2</sub> is necessary for the urea synthesis. In Linz, near to one of the main sites for iron and steel production, a urea synthesis plant with a production rate of around 400,000 t per year of urea is located [46]. Therein, around 300,000 t CO<sub>2</sub> per year are required for the production of the given amount of urea [46]. Further utilization possibilities could be CO2-derived fuels, like methanol or FTsynthesis and power to gas. Furthermore, the utilization within CO<sub>2</sub>-derived chemicals beside urea, like formic acid synthesis, or CO<sub>2</sub>-derived building materials, like the production of concrete, could be promising alternatives [45].

Beside the CCU technologies,  $CO_2$  can also be stored in underground deposits. CCS is banned in Austria except research projects up to a storage volume of 100,000 t of  $CO_2$ [44]. For further information regarding CCU and CCS, a reference is made to [40, 45, 47–49].

Since biomass releases the same amount of  $CO_2$  as it aggregates during its growth, the utilization of biogenic fuels can contribute significantly to a reduction of  $CO_2$  emissions. Therefore, the main focus of the paper lies on the production of a below zero emission reducing gas by the use of oxyfuel combustion in combination with sorption-enhanced reforming. This technology for the selective separation of  $CO_2$  uses as fluidization agent a mix of pure oxygen and recirculated flue gas. Therefore, the nitrogen from the air is excluded from the combustion system [42].

### 2.2 Combination of oxyfuel combustion and sorptionenhanced reforming

A promising option for the selective separation of  $CO_2$  from biomass and the generation of a hydrogen-rich product gas at the same time is the sorption-enhanced reforming process in combination with oxyfuel combustion (OxySER). The sorption-enhanced reforming (SER) is based on the dual fluidized bed steam gasification process. The main carbonrelated (gas-solid) and gas-gas reactions are shown in Table 2. Test runs at the 100 kW pilot plant at TU Wien showed calculated overall cold gas efficiencies of around 70% [51, 52]. Detailed information regarding the dual fluidized bed steam gasification process can be found in literature [37, 51–54].

The combination of oxyfuel combustion and sorptionenhanced reforming combines the advantages of both technologies. Figure 1 represents the concept of the combined technology [44]. First of all, biomass, residues, or waste materials are introduced in the gasification reactor. Limestone is used as bed material which serves as transport medium for heat but also as carrier for CO<sub>2</sub> from the gasification reactor (GR) to the combustion reactor (CR) by adjusting the temperature levels in the reactors correctly. Within the OxySER process, steam serves as fluidization and gasification agent in the GR. Therein, several endothermic gasification reactions take place in a temperature range between 600 and 700 °C [37]. Residual char is transferred with the bed material from the GR to the CR. Due to the combination of SER with oxyfuel combustion, pure oxygen instead of air is used as fluidization agent in the CR, which is operated within a temperature range between 900 and 950 °C. By combustion of residual char in the CR, heat is released. This suitable temperature profiles in the GR and CR ensure that the bed material (limestone) is first calcined to calcium oxide (CaO) at high temperatures in the CR (13). Then the CaO is carbonized in the GR with the carbon dioxide from the product gas (12). Thus, in this cyclic process, a transport of CO<sub>2</sub> from the product gas to the flue gas appears [52]. The use of steam in the gasification reactor and the water gas shift reaction (8) in combination with in situ  $CO_2$  sorption via the bed material system CaO/CaCO<sub>3</sub> enables the production of a nitrogen-free and hydrogen-enriched product gas [37, 56]. Due to the combination of SER with oxyfuel combustion, in addition to the nitrogen-free and hydrogen-enriched product gas, a CO<sub>2</sub>-enriched flue gas is generated caused by the use of pure oxygen as fluidization agent in the CR instead of air [57].

The  $CO_2$  equilibrium partial pressure in the CaO/CaCO<sub>3</sub> system and the associated operation conditions for the

gasification and combustion can be found in [52]. By the use of renewable fuels and a continuous selective separation and storage or utilization of  $CO_2$ , an improved  $CO_2$  balance can be achieved [44, 57].

Table 3 represents a comparison between the product and flue gas compositions of conventional gasification, SER, and OxySER. The results are based on test runs with the 100 kW pilot plant at TU Wien and the 200 kW pilot plant at University of Stuttgart [37, 57]. As mentioned above, the carbon dioxide content of the product gas could be reduced through the SER method. Furthermore, the hydrogen content is higher in comparison with conventional gasification. The possibility of adjusting the H<sub>2</sub>/CO ratio over a wide range makes the SER process very flexible according to product gas applications [52]. The catalytic activity of limestone enables a reduction of tar at the same time [37, 44, 58]. The comparison between the SER and OxySER process illustrates that a CO<sub>2</sub>-enriched flue gas in the OxySER test rig in Stuttgart was obtained [57]. In Table 4 the proximate and ultimate analyses of used wood pellets for gasification test runs with the 100 kW pilot plant at TU Wien are listed.

However, OxySER implies the following advantages in comparison to the conventional gasification:

- Selective CO<sub>2</sub> transport to flue gas
- Decrease of tar content in product gas
- High CO<sub>2</sub> content in flue gas > 90 vol.- $\%_{dry}$  [57]
- · Smaller flue gas stream because of flue gas recirculation
- Nitrogen free flue gas

These assumptions according to experimental results serve as a basis for the conception of an industrial application.

# 2.3 Integrated OxySER concept for the production of below zero emission reducing gas

The OxySER plant concept for integration in a direct reduction plant is illustrated in Fig. 2. The plant concept is designed for a product gas power of 100 MW. For the production of 100 MW product gas, 50,400 kg/h of wood chips with a water content of 40 wt.-% are required [37]. The wood chips are treated in a biomass dryer. Afterwards the biomass is fed in the gasification reactor. The bed material inventory (limestone) of the system contains 25,000 kg. In the gasification reactor, a H<sub>2</sub>-enriched product gas with a temperature of 680 °C is produced. Subsequently, the dust particles are removed from the product gas by a cyclone. Besides ash, these dust particles contain still carbon. This is the reason why the particles are recirculated to the combustion reactor. Afterwards, the product gas is cooled down to 180 °C. The released heat can be used for preheating of the biomass dryer air [44]. Furthermore, the product gas filter separates further fine dust particles from the product gas stream and conveys them back to the combustion reactor. After that, tar is separated in a scrubber, and water is condensed. Biodiesel (RME) is used as solvent. The product gas exits the scrubber with a temperature of 40 °C. Afterwards, it is compressed in a blower, before it is dried to a water content of 1.5% and fed to the compression and preheating of the direct reduction plant. The CO<sub>2</sub>-enriched flue gas leaves the combustion reactor with a temperature of 900 °C. The flue gas is cooled down to 180 °C by the steam superheater and a flue gas cooler. Steam is heated up to 450 °C in a countercurrent heat exchanger. Fly ash is removed out of the system by a flue gas filter. A partial flow from the flue gas is recirculated and mixed with pure oxygen. Pure oxygen is produced by an air separation unit. The remaining flue gas stream is compressed in the flue gas blower, and water is condensed in a flue gas dryer. The cleaned CO<sub>2</sub>rich gas can be used in different CCU processes, like urea or methanol synthesis [44].

The integration approach offers the advantage to use existing equipment, like the air separation unit from the steelmaking facility. Furthermore, the generated product gas can be used directly in the direct reduction plant, as reducing gas [44]. For this application, a compression up to approx. 2.5 bar and preheating of the product gas up to 900 °C are necessary.

# 2.4 Simulation of mass and energy balances with IPSEpro

The calculation of mass and energy balances for different operation points with the stationary equation-orientated flow sheet simulation software IPSEpro enables the validation of process data. All data which cannot be measured during experimental test runs can be determined by the calculation of closed mass and energy balances. These equations are solved by the numerical Newton-Raphson Algorithm [59, 60]. Therefore, no models regarding kinetic or fluid dynamic approaches are considered. The used simulation models within the software IPSEpro are based on model libraries, which were developed at TU Wien over many years [61]. All experimental results from the pilot plant at TU Wien, presented within this publication, were validated with IPSEpro. Uncertainties are given by the accuracy of measurement data which relies on used analysis methods. The measurement accuracy of the ultimate and proximate analysis is listed in Table 4. The validation percentage error of the gasification model is covered by the range of values which are listed in Table 3. For further information regarding IPSEpro, a reference is made to [61, 62]. Due to the validation of the results from the pilot plant at University of Stuttgart, a reference is made to [57].

The simulation results for the OxySER concept for the production of below zero emission reducing gas presented in Section 2.3 are based on scale up of the experimental results of the pilot plants. The simulation model of the dual fluidized

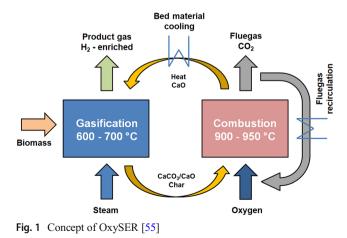
#### Table 2 Important gas-solid and gas-gas reactions during thermochemical fuel conversion [50]

Important heterogeneous reactions (gas-solid)			
Oxidation of carbon	$C + O_2 \rightarrow CO_2$	Highly exothermic	(1)
Partial oxidation of carbon	$C + \frac{1}{2} O_2 \rightarrow CO$	Exothermic	(2)
Heterogeneous water-gas shift reaction	$C + H_2 O \to CO + H_2$	Endothermic	(3)
Boudouard reaction	$C + CO_2 \rightarrow 2 CO$	Endothermic	(4)
Hydrogenation of carbon	$C + 2 H_2 \rightarrow CH_4$	Slightly exothermic	(5)
Generalized steam gasification of solid fuel (bulk reaction)	$C_x H_y O_z + (x-z)H_2 O \rightarrow x CO + (x-z+\frac{y}{2})H_2$	Endothermic	(6)
Important homogeneous reactions (gas-gas)			
Oxidation of hydrogen	$2 H_2 + O_2 \rightarrow 2 H_2O$	Highly exothermic	(7)
Homogeneous water-gas shift reaction	$CO + H_2O \rightarrow CO_2 + H_2$	Slightly exothermic	(8)
Methanation	$CO + 3 H_2 \rightarrow CH_4 + H_2O$	Exothermic	(9)
Generalized steam reforming of hydrocarbons	$C_x H_y + x H_2 O \rightarrow x CO + (x + \frac{y}{2}) H_2$	Endothermic	(10)
Generalized dry reforming of hydrocarbons	$C_x H_y + x CO_2 \rightarrow 2x CO + \frac{y}{2} H_2$	Endothermic	(11)
Important reactions of active bed material (limestone) for SER			
Carbonation	$CaO + CO_2 \rightarrow CaCO_3$	Exothermic	(12)
Calcination	$CaCO_3 \rightarrow CaO + CO_2$	Endothermic	(13)

bed steam gasification system is based on an exergy study of T. Pröll [63].

# 2.5 Techno-economic assessment with net present value calculation

The techno-economic assessment regarding the net present value (NPV) calculation serves as decision-making tool for the valuation of upcoming investments. The NPV is a function of the investment and operating costs. The operating costs are multiplied by the cumulative present value factor, which includes the interest rate and the plant lifetime. Therefore, the NPV calculation helps to compare expected payments in the future with current payments. Further information can be found in [54, 64]. Cost rates have been updated to the year 2019 by using data from a chemical engineering plant cost index (CEPCI) database [65]. For the calculation of the investment costs, the cost-scaling method was used [66].



The techno-economic analysis is based on the following *business case* that an operator of a direct reduced iron plant would like to build a new reducing gas supply unit driven by a biogenic feedstock. The goal to produce 100 MW reducing gas should be achieved with regard to  $CO_2$  emissions. The reference option (option 0) is the production of reducing gas by steam reforming of natural gas. Furthermore, three biogenic alternative options (options 1–3) are compared with the reference option:

- *Option 0* (reference case): Production of 100 MW reducing gas through steam reforming of natural gas
- Option 1: Production of 100 MW reducing gas through gasification of wood chips by SER
- Option 2: Production of 100 MW reducing gas through gasification of wood chips by an integrated OxySER plant
- Option 3: Production of 100 MW reducing gas through gasification of wood chips by a greenfield OxySER plant

The SER process in option 1 requires no pure oxygen, consequently no ASU for operation. However, the flue gas of the SER process cannot be exploited in further utilization steps because of the high nitrogen content in the flue gas. The alternative option 2 is based on the SER process in combination with oxyfuel combustion implemented in an existing iron and steel plant facility. The process heat is used for preheating of the reducing gas. The required oxygen is delivered from an existing ASU within the iron and steel plant facility. Furthermore, the OxySER process is based on the assumption that the  $CO_2$  is sold as product for utilization to a urea synthesis plant. Option 3 is based on the OxySER process without the benefits from option 2.

Parameter	Unit	Conventional gasification (100 kW)	Gasification by SER (100 kW)	Gasification by SER (200 kW)	Gasification by OxySER (200 kW)
Plant location		TU Wien	TU Wien	University Stuttgart	University Stuttgart
Reference		[37]	[37]	[57]	[57]
Fuel		Wood pellets	Wood pellets	Wood pellets	Wood pellets
Bed material		Olivine	Limestone	Limestone	Limestone
Particle size	mm	0.4–0.6	0.5-1.3	0.3-0.7	0.3-0.7
Product gas composition					
Water (H <sub>2</sub> O)	vol%	30–45	50-65	50	50
Hydrogen (H <sub>2</sub> )	vol% <sub>dry</sub>	36–42	55–75	69–72	70
Carbon monoxide (CO)	vol%dry	19–24	4–11	8-11	8
Carbon dioxide (CO <sub>2</sub> )	vol%dry	20–25	6–20	5–7	8
Methane (CH <sub>4</sub> )	vol% <sub>dry</sub>	9–12	8-14	11-12	11
Non cond. hydrocarbons $(C_xH_y)$	vol% <sub>dry</sub>	2.3–3.2	1.5–3.8	2–3	3
Dust particles	g/Nm <sup>3</sup>	10–20	20–50	n.m.	n.m.
Tar	g/Nm <sup>3</sup>	4-8	0.3–0.9	14	6
Flue gas composition					
Water (H <sub>2</sub> O)	vol%	n.m.	n.m.	14	30
Oxygen (O <sub>2</sub> )	vol% <sub>dry</sub>	n.m.	n.m.	7	9
Nitrogen (N <sub>2</sub> )	vol% <sub>dry</sub>	n.m.	n.m.	46	-
Carbon dioxide (CO <sub>2</sub> )	vol% <sub>dry</sub>	n.m.	n.m.	47	91

 Table 3
 Comparison product and flue gas composition of conventional gasification, SER, and OxySER [37, 57]

n.m., not measured

This means that, in option 3, the costs for pure oxygen are higher in consideration to the use of a greenfield ASU. Furthermore, no earnings through  $CO_2$  utilization are considered.

$$A = P^* \frac{\mathrm{IR}^* (1 + \mathrm{IR})^n}{(1 + \mathrm{IR})^n - 1}$$

Furthermore, a payback analysis has been done by solving the following equation, where A are the savings minus the operation and maintenance costs, P is the present worth capital costs, and IR is the interest rate. The variable n represents the number of years to return the investment in comparison with the reference case [67].

# **3** Results and discussion

Based on experiences of the pilot plant from the TU Wien and the University of Stuttgart, combined with the previously

 Table 4
 Proximate and ultimate analyses of used wood pellets for gasification test runs [51]

Parameter	Unit	Meas. accuracy (%)	Wood pellets (100 kW)
Water content (H <sub>2</sub> O)	wt%	±4.3	7.2
Ash content (550 °C)	wt% <sub>dry</sub>	$\pm 9.2$	0.2
Carbon (C)	wt% <sub>daf</sub>	$\pm 1.0$	50.8
Hydrogen (H)	wt% <sub>daf</sub>	$\pm 5.0$	5.9
Nitrogen (N)	wt% <sub>daf</sub>	$\pm 5.0$	0.2
Sulfur (S)	wt% <sub>daf</sub>	±7.5	0.005
Chlorine (Cl)	wt% <sub>daf</sub>	$\pm 7.5$	0.005
Oxygen (O) <sup>*</sup>	wt% <sub>daf</sub>	-	43.1
Volatile matter	wt% <sub>daf</sub>	$\pm 0.45$	85.6
Lower heating value, moist	MJ/kg	$\pm 1.0$	17.4

\* Calculated by difference to 100 wt.-%daf

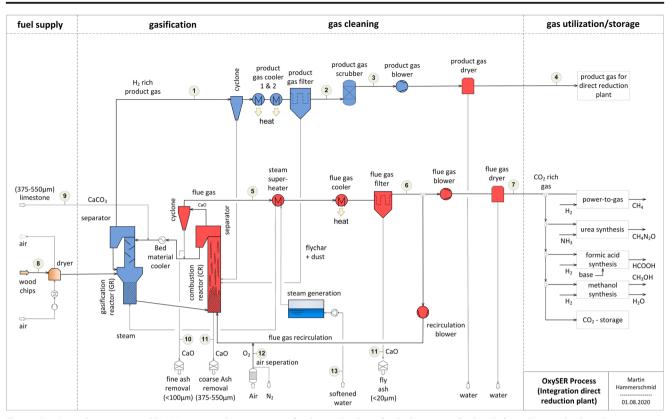


Fig. 2 OxySER plant concept with 100-MW product gas power for the production of reducing gas as feedstock for a direct reduction plant

described concept, mass and energy balances for the OxySER plant concept for integration in a direct reduction plant were calculated. Furthermore, mass and energy balances are the basis for a techno-economic assessment. In Table 5 the most important streamline data of chosen flow streams, marked in Fig. 2, are shown. Table 6 and Table 7 represent the input and output data and operating parameters of an OxySER plant.

Table 6 shows the input and output flows of an OxySER plant with 100 MW product gas energy. It can be seen that 50,400 kg/h of wood chips and 11,020 Nm<sup>3</sup>/h of pure oxygen are required for the generation of 28,800 Nm<sup>3</sup>/h product gas. The product gas is used as reducing gas in the direct reduction route. Furthermore, 36,100 kg/h of CO<sub>2</sub> can be recovered for further utilization. The costs for final disposal of 1050 kg/h of ash and dust have been taken into account.

In Table 8, the main requirements on the product gas for the utilization in the direct reduction plant are listed. The comparison illustrates that the generated below zero emission product gas out of the OxySER plant meets, except from the temperature and pressure, all the requirements. The concept is based on the assumption that the reducing gas is compressed and preheated before it is fed to the direct reduction plant. Therefore, the required temperature and pressure are reached after compression and preheating of the product gas.

The techno-economic assessment relies on the results of the IPSEpro simulation. Table 9 represents the fuel prices for chosen fuel types and cost rates for utilities. It is thus evident that the European natural gas price with 25  $\epsilon$ /MWh is more expensive than in other continents. Exemplary, the costs for one employee per year are assumed to 70,000  $\epsilon$ /a, and the expected plant lifetime of an OxySER plant is 20 years.

Table 10 represents the investment cost rates for the NPV calculation. The presented investment costs are based on total capital investment costs of realized fluidized bed steam gasification plants driven as combined heat and power plants reduced by the costs through the gas engine. Furthermore, this investment costs are updated by CEPCI and scaled with the cost-scaling method. For the integrated OxySER plant, the assumption was made that the oxygen from the air separation unit (ASU) of the iron and steel plant is used. For the green-field OxySER plant, the whole investment costs for an ASU were added.

The techno-economic analysis is based on the Section 2.5 that described *business case*, wherein an operator of a direct reduced iron plant would like to build a new reducing gas supply unit driven by a biogenic feedstock. The NPV calculation, which is shown in Table 11, serves as decision-making tool. The goal to produce 100 MW reducing gas should be achieved with regard to  $CO_2$  emissions. The reference option (option 0) is the production of reducing gas by steam reforming of natural gas. Furthermore, three biogenic alternative options (options 1–3), which are described in Section 2.5, are compared with the reference option.

Parameter	Unit	Product gas st	reams			Flue gas str	reams	
		Product gas after GR	Product gas after filter	Product gas after scrubber	Reducing gas for DR	Flue gas after CR	Flue gas after filter	CCU
Streamline in Fig. 2	-	(1)	(2)	(3)	(4)	(5)	(6)	(7)
Pressure	Bara	Ambient	Ambient	Ambient	Ambient	Ambient	Ambient	Ambient
Temperature	°C	675	150	40	60	950	160	160
Mass flow rate	kg/h	26,000	25,500	16,000	15,800	93,200	92,600	36,100
Volume flow rate	Nm <sup>3</sup> /h	40,500	40,000	28,800	28,400	53,000	52,500	20,500
Water content	wt%	35.0	35.0	8.0	1.5	15.0	15.0	5.0
Hydrogen (H <sub>2</sub> )	vol% <sub>dry</sub>	69.2	69.2	69.2	69.2	0	0	0
Carbon monoxide (CO)	vol% <sub>dry</sub>	9.1	9.1	9.1	9.1	2.8	2.8	2.8
Carbon dioxide (CO <sub>2</sub> )	vol% <sub>dry</sub>	6.5	6.5	6.5	6.5	91.2	91.2	91.2
Methane (CH <sub>4</sub> )	vol% <sub>dry</sub>	11.0	11.0	11.0	11.0	0	0	0
Non cond. Hydrocarbons (C <sub>x</sub> H <sub>y</sub> )	vol% <sub>dry</sub>	2.4	2.4	2.4	2.4	0	0	0
Oxygen $(O_2)$	vol% <sub>dry</sub>	0.1	0.1	0.1	0.1	6.0	6.0	6.0
Nitrogen (N <sub>2</sub> )	vol%dry	1.7	1.7	1.7	1.7	0	0	0
Dust particle	g/Nm <sup>3</sup>	10	0.025	0	0	20	0	0
Tar content*	g/Nm <sup>3</sup>	4	0.5	0.025	0.025	0	0	0

 Table 5
 Streamline data of the OxySER concept according to Fig. 2

\*Tar is considered in the simulation model as naphthalene (main component in the DFB product gas) [51]

Table 11 represents the net present value calculation for the production of 100 MW reducing gas. Therein, the fuel energy per year, the investment costs including interest and fuel costs per year are listed. Beside the fuel costs, Table 11 shows also all other consumption-related costs. Costs for  $CO_2$  emission certificates are paid only for the use of fossil fuels (reference case). The relative NPV represents the profitability of alternative production routes in comparison with the reference case and the payback period for return of investment. The NPV of all alternative options (1-3) shows negative values. This means that the operation of SER and OxySER with wood chips based on the expected plant lifetime of 20 years is less profitable than the reference option. The techno-economic comparison between SER and OxySER shows that in option 2, the earnings through carbon dioxide are higher than the oxygen costs. In option 3, no earnings through CO<sub>2</sub> utilization and no benefits regarding oxygen costs have been considered. Therefore, an extremely negative NPV in option 3 is the result. The payback analysis shows that only option 2

 Table 6
 Input and Output data of an OxySER plant with 100 MW product gas energy

Input					Output				
Parameter	Streamline in Fig. 2	Unit	Value	Ref.	Parameter	Streamline in Fig. 2	Unit	Value	Ref.
Bed material inventory	-	kg	25,000	[37]	Product gas	(3)	Nm <sup>3</sup> /h	28,800	IPSE
Fuel (wood chips)	(8)	kg/h	50,400	[37]	Flue gas	(8)	Nm <sup>3</sup> /h	53,000	IPSE
Fresh bed material	(9)	kg/h	1770	[37, 64]	Ash and dust	(11)	kg/h	1050	[37]
Cooling capacity in % of fuel power	_	% (kW/kW <sub>th</sub> )	5–20	[68]	Bed material	(10)	kg/h	1000	[44]
Electricity consumption	_	kW	2800	[37]	Carbon dioxide (for CCU)	(7)	kg/h	36,100	[37]
Oxygen	(12)	Nm <sup>3</sup> /h	11,020	[37]					
Fresh water	(13)	kg/h	378	[37]					
Scrubber solvent (RME)	-	kg/h	200	[37]					
Flushing gas	-	Nm <sup>3</sup> /h	500	[37]					

**Table 7**Operating parameters of an OxySER plant with 100 MWproduct gas energy

Parameter	Unit	Value	Ref.
Lower heating value, moist (wood chips)	MJ/kg	9.53	[37]
Water content (wood chips)	wt%	40	[37]
Combustion temperature	°C	900–950	[ <mark>69</mark> ]
Gasification temperature	°C	625–680	[ <mark>69</mark> ]
Particle size (bed material)	μm	375-550	Asm.
Coarse ash	μm	375-550	Asm.
Fine ash	μm	< 100	Asm.
Very fine ash	μm	< 20	Asm.
Water content (PG to DR)	vol%	1.50	IPSE
CO <sub>2</sub> recovery rate <sup>*</sup>	%	>95	IPSE

\*CO<sub>2</sub> =  $\frac{CO_2 \text{ volume flow flue gas}}{CO_2 \text{ volume flow total } (FG+PG)}$ 

could return the investment regarding the expected interest rate in comparison with the reference case. However, the payback time of 24 years is very long and would not be profitable. Option 1 and option 3 could not return the investment in comparison to the reference case.

Furthermore, the reducing gas production costs of the four different routes were calculated. As can be seen from Table 11, the production costs (LCOP) of the reference case are with 39.0 €/MWh as the lowest followed by the integrated OxySER process with 39.4 €/MWh. Figure 3 represents the discounted expenses and revenues, divided in the main cost categories. It can be seen that the fuel costs are the main cost driver in the process. The techno-economic comparison points out that the production costs of a below zero emission reducing gas could only be in the range of steam-reformed natural gas, if generated  $CO_2$  can be utilized and the pure oxygen is delivered by an integrated ASU. Otherwise, the production of biomass-based reducing gas via the SER process is preferable. A further reduction of the production costs of the biomassbased reducing gas could be reached by the use of cheaper fuels.

 Table 8
 Requirements on product gas for the utilization in the direct reduction plant [22, 70]

Parameter	Unit	Requirement reducing gas	Value product gas
Temperature	°C	> 900	60
Pressure	bara	2–4	1.05
H <sub>2</sub> /CO ratio	-	0.5 - ∞	7.6
Gas quality*	-	>9	9.8
Methane	vol%	> 3.5	11.0
Sulfur $(H_2S)$	ppm	< 100	< 20
Soot	mg/Nm <sup>3</sup>	< 100	-

\*Gas quality =  $(\%CO + \% H_2)/(\%CO_2 + \% H_2O)$  [70]

Additionally, a sensitivity analysis of the NPV calculation has been created. The results for the sensitivity analysis based on the NPV of option 2 are shown in Fig. 4. The sensitivity analysis shows that the fuel prices of natural gas and wood chips are the most sensitive cost rates. The fuel cost rates depend very much on the plant location. Furthermore, the NPV in this techno-economic comparison is also sensitive to the investment costs of the reducing agent production route, the revenues through CCU, the price of CO<sub>2</sub> emission certificates, the plant lifetime, the operating hours, and the interest rate. The revenues through CCU depend on the availability of consumers. The sensitivity to operating hours and plant life time reaffirms high importance to a high plant availability during the whole plant life cycle. Cost rates for operating utilities, maintenance, and employees are less sensible to the results.

Finally, a comparison of the production costs of the biomass-based reducing gas with other reducing agents like reformed natural gas, hydrogen, or coke has been done. The comparison in Fig. 5 shows that the production of biomassbased reducing gas via OxySER (option 2) and SER is more than twice as expensive as the production of coke in a coking plant, but it is in the same range than the production of reducing gas via steam reforming of natural gas. All fuel costs are based on European price levels. Especially, the natural gas price strongly depends on the plant site. For example, the natural gas price in Europe is four to five times higher than in North America [33]. This is the reason why most of the existing direct reduction plants are built in oil-rich countries [33]. The production of hydrogen using water electrolysis is currently economically not competitive. On the ecologic point of view, the use of biomass-based reducing gas without CCU decrease the CO<sub>2</sub> emissions of the whole process chain for the production of crude steel down to 0.28 t  $CO_2e/t_{CS}$ . This amounts to a reduction of CO<sub>2</sub> emissions in comparison with the integrated BF-BOF route by more than 80%. Further on, the use of CCU within an OxySER plant could create a CO2 sink, since biomass releases the same amount of  $CO_2$  as it aggregates during its growth.

With regard to 8.1 million tons of crude steel production in Austria, in the year 2017 [4], and an estimated woody biomass potential of around 50 PJ in the year 2030 [21], 13 biomass-based reducing gas plants (OxySER or SER) with a reducing gas power of 100 MW could be implemented. This would result in the production of around 35 Mio. GJ of biomass-based reducing gas for the direct reduction process, which is sufficient for the production of 3.5 Mio. tons of crude steel. One of the biomass-based reducing gas plants could be operated via the OxySER process with regard to the CCU potential from the nearby urea synthesis plant of 300,000 t CO<sub>2</sub> per year [46]. Further CCU potential could be arise through the production of CO<sub>2</sub>-derived fuels or chemicals [41].

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Utility cost rate	Unit	Value	Ref.	NPV cost rate	Unit	Value	Ref.
Wood chips (Austria)	€/MWh	15.7	[71]	Maintenance costs per year	%/a	2.00	[54]
Natural gas (Austria)	€/MWh	25.0	[72]	Insurance, administration, and tax per year	%/a	1.50	[73]
Electricity	€/kWh <sub>el</sub>	0.04	[64]	Number of employees (integration)	-	3	[64]
Limestone	€/t	35	[64]	Number of employees (greenfield)	-	7	[44, 64]
Nitrogen	€/Nm <sup>3</sup>	0.003	[64]	Expected plant life time	а	20	[73]
Fresh water	€/t	0.02	[64]	Annual operating hours	h/a	7500	[64]
Solvent (RME)	€/t	960	[64]	Interest rate (IR)	%	6	[74]
Oxygen (air separator available)	€/Nm <sup>3</sup>	$0.022^*$	[44]	Costs of one employee per year	€/a	$70,000^{*}$	[64]
Oxygen (greenfield)	€/Nm <sup>3</sup>	$0.075^{*}$	[37]				
Emission allowances certificate	€/t <sub>CO2</sub>	23	[75]				
Costs for ash disposal	€/t	90	CHP Güssing				
$CO_2$ expenses	€/Nm <sup>3</sup>	0.03	[76]				

Table 9 Cost rates for utilities and NPV calculation

# 4 Conclusion and outlook

The scope of this publication was the investigation of a concept for the production of a below zero emission reducing gas for the use in a direct reduction plant and whether it has a reasonable contribution to a reduction of fossil CO<sub>2</sub> emissions within the iron and steel sector in Austria. The gasification via SER allows the in situ CO<sub>2</sub> sorption via the bed material system CaO/CaCO<sub>3</sub>. Therefore, a selective transport of carbon dioxide from the product gas to the flue gas stream is reached. The use of a mix of pure oxygen and recirculated flue gas as fluidization agent in the CR results in a nearly pure CO<sub>2</sub> flue gas stream. Through the in situ CO<sub>2</sub> sorption, CO<sub>2</sub> recovery rates up to 95% can be reached. The CO<sub>2</sub> could be used for further synthesis processes like, e.g., the urea synthesis. Therefore, a below zero emission reducing gas could be produced.

The experimental and simulation results show that the produced below zero emission OxySER product gas meets all requirements for the use in a direct reduction plant. The use of the biomass-based reducing gas out of the SER process within a MIDREX plant would decrease the emitted CO<sub>2</sub> emission by 83% in comparison to the blast furnace route. The use of a below zero emission reducing gas out of the OxySER process by the use of CCU would create a CO<sub>2</sub> sink. The results of the techno-economic assessment show that the production of reducing gas via sorption-enhanced reforming in combination with oxyfuel combustion can compete with the natural gas route, if the required pure oxygen is delivered by an available ASU and if CCU is possible. Otherwise, the SER process is more profitable. Furthermore, the sensitivity analysis of the cost rates exhibited that the fuel and investment costs are strongly dependent on the profitability of the OxySER plant and in consequence the direct reduction plant.

Table 10	Investment	costs fe	or NPV	calculation

Parameter	Unit	Value	Ref.
Investment costs SER plant (total capital investment costs)*	Mio. €	85	[37] adapted by CEPCI
Investment costs integrated OxySER plant (SER plus maintenance ASU)**	Mio. €	91	[37, 66] adapted by CEPCI
Investment costs greenfield OxySER plant (SER plus total investment costs ASU)***	Mio. €	115	[37, 66] adapted by CEPCI
Investment costs Steam Reformer natural gas	Mio. €	54	[77] adapted by CEPCI

\*Investment costs are based on scaled total capital investment costs of realized dual fluidized bed steam gasification plants driven as combined heat and power plants reduced by the costs of the gas engine/investment costs updated with CEPCI [37]

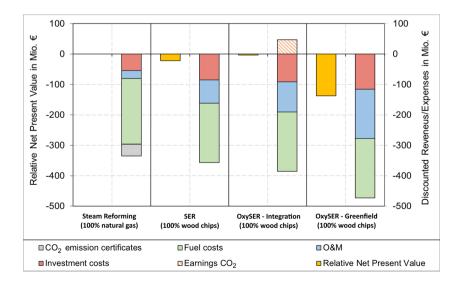
\*\*Investment costs are based on costs SER plant raised by a third of the ASU maintenance costs (2% of the investment costs per year with an expected lifetime of 20 years)/assumption: 50% of ASU is used for OxySER plant and 50% for iron and steel plant

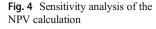
\*\*\*ASU investment costs: approx. 30 Mio. € [66] adapted by CEPCI

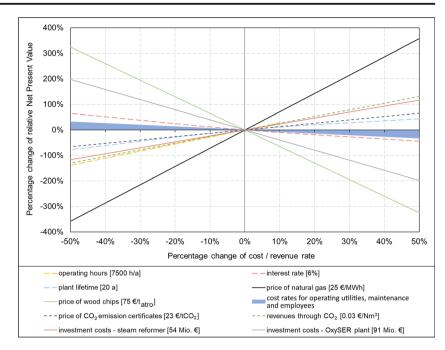
### Table 11 Net present value calculation for the production of 100 MW reducing gas

Parameter	Unit	Steam Reforming (100% natural gas) Option 0	SER (100% wood chips) Option 1	Integration OxySER (100% wood chips) Option 2	Greenfield OxySER (100% wood chips) Option 3			
Boundary conditions								
Reducing gas for direct reduction	MW	100	100	100	100			
Natural gas consumption	MWh/a	750 000						
Wood chips consumption	MWh/a		997 500	997 500	997 500			
Investment costs incl. interest	€	54 000 000	85 000 000	91 000 000	115 000 000			
Expenses								
Fuel costs natural gas	€/a	18 750 000						
Fuel costs wood chips	€/a		17 010 000	17 010 000	17 010 000			
CO <sub>2</sub> emission certificates	€/a	3 450 000						
Maintenance, insurance, etc.	€/a	1 890 000	2 975 000	3 185 000	4 025 000			
Employee costs	€/a	70 000	210 000	210 000	490 000			
Auxiliaries	€/a	356 000	1 916 000	1 916 000	1 916 000			
Electricity costs	€/a	556 000	835 500	835 500	835 500			
Ash disposal costs	€/a		709 000	709 000	709 000			
Oxygen costs	€/a			1 818 000	6 198 500			
Sum of expenses per year	€/a	24 516 000	23 655 500	25 683 500	31 184 000			
Earnings								
Earnings CO <sub>2</sub> utilization	€/a			4 102 000				
Sum of earnings per year	€/a			4 102 000				
Net present value calculation								
Expenses - Earnings	€/a	24 516 000	23 655 500	21 581 500	31 184 000			
Additional investment costs (P) (compared to reference option)	€	0	31 000 000	37 000 000	61 000 000			
Operating expenses savings (A)	€/a	0	860 500	2 934 500	- 6 668 000			
<b>Relative Net Present Value</b>	€	0	- 21 128 000	- 3 340 000	- 137 500 000			
Payback analysis (Return of investment period compared to reference case)								
Payback time (n)*	а	-	$\infty$	24	8			
Production costs reducing gas (LCOP)								
Production costs reducing gas	€/MWh	39.0	41.4	39.4	54.9			
(LCOP)**	€/GJ	10.8	11.5	10.9	15.3			
* Payback analysis: $A = P * \frac{IR^*(1+IR)^n}{(1+IR)^{n-1}} [67]$								
** LCOP = $\frac{Sum \ of \ discounted \ (expenses-earnings)}{Discounted \ Delivered \ reducing \ gas} = \frac{\epsilon}{MWh} = \frac{\epsilon}{GJ} [78, 79]$								

Fig. 3 Relative net present value

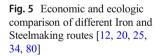


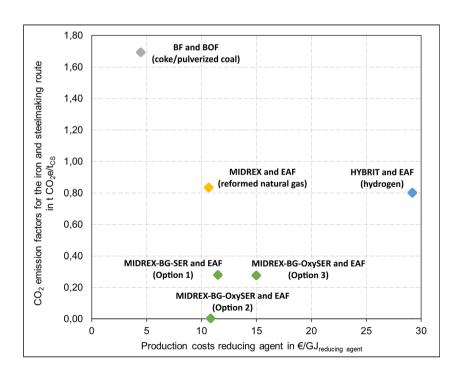




The production costs of the biomass-based reducing gas are more than twice as high as the fossil coke, which is used mainly in the blast furnace route.

Summing up, the presented integrated concept and the calculated results enable valuable data for further design of the proposed concept. Beforehand a demonstration at a significant scale is recommended. Further on, the implementation of the energy flows from an iron and steel plant within the simulation model could improve the current model regarding to efficiency. The profitability of the direct reduction with a biomassbased reducing gas or natural gas is strongly dependent on the availability of sufficient fuel. With regard to the woody biomass potentials in Austria in the year 2030, the production of 3.5 Mio. tons of crude steel by the use of biomass-based reducing gas could be reached. Due to the substitution of the integrated BF and BOF route by the MIDREX-BG-SER and EAF route, the reduction of 6.8 Mio. tons of CO<sub>2</sub>e could be reached. This amount would decrease the CO<sub>2</sub> emissions within the iron and steel sector in Austria by 50%. Concluding, the production of biomass-based reducing gas





could definitely help to contribute on the way to defossilization of the iron and steelmaking industry in Austria.

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**Data availability** The data that support the findings of this study are available from the corresponding author, M. Hammerschmid, upon reasonable request.

#### Compliance with ethical standards

**Conflicts of interest** The authors declare that they have no conflict of interest.

Code availability Not applicable.

Abbreviations AISI, American Iron and Steel Institute; Asm., assumption; ASU, air separation unit; BF, blast furnace; BG, biomass-based reducing gas; BOF, basic oxygen furnace; C, carbon; CaCO<sub>3</sub>, calcium carbonate; CaO, calcium oxide; CCS, carbon capture and storage; CCS/ U, carbon capture and storage or utilization; CCU, carbon capture and utilization; CEPCI, chemical engineering plant cost index; CH<sub>3</sub>OH, methanol; CH<sub>4</sub>, methane; CH<sub>4</sub>N<sub>2</sub>O, urea; CHP, combined heat and power; CIRCORED, novel direct reduction technology; CO, carbon monoxide; CO2, carbon dioxide; CO2e, carbon dioxide equivalent; COG, coke oven gas; COREX, smelting reduction technology; COURSE50, CO2 ultimate reduction steelmaking process by innovative technology for cool Earth 50 located in Japan; CR, combustion reactor; CS, crude steel; CxHv, non condensable hydrocarbons; DR, direct reduction; DRI, direct reduced iron; dry, dry basis; EAF, electric arc furnace; EU-28, member states of the European Union (until January 2020); FG, flue gas; FINEX, smelting reduction technology; FINMET, direct reduction technology; GR, gasification reactor; H<sub>2</sub>, hydrogen; H<sub>2</sub>O, water; H<sub>2</sub>S, hydrogen sulfide; HCOOH, formic acid; HISARNA, novel bath-smelting technology; HM, hot metal; HYBRIT, Hydrogen Breakthrough Ironmaking Technology; IPSEpro, software tool for process simulation; LCOP, levelized costs of products; MIDREX, state-of-the-art direct reduction technology; MOE, molten oxide electrolysis; N2, nitrogen; NH3, ammonia; NPV, net present value; O2, oxygen; OPEX, operational expenditure; OxySER, sorption-enhanced reforming in comb. with oxyfuel combustion; PG, product gas; POSCO, iron and steelmaking company located in Korea; Ref., reference; RME, rapeseed methyl ester; SER, sorption-enhanced reforming; t<sub>CS</sub>, tons of crude steel; TGR, top-gas recycling; ULCOLYSIS, novel electric direct reduction technology; ULCORED, novel direct reduction technology; ULCOS, ultra-low CO2 steelmaking; ULCOWIN, novel electric direct reduction technology; vol.-%, volumetric percent wet; vol.-%dry, volumetric percent dry; wt.-%, weight percent wet; wt.-%daf, weight percent dry and ash free; wt.-%dry, weight percent dry

Symbols %CO, volume percent of carbon monoxide within reducing gas; %CO<sub>2</sub>, volume percent of carbon dioxide within reducing gas; %H<sub>2</sub>, volume percent of hydrogen within reducing gas; %H<sub>2</sub>O, volume percent of water within reducing gas; A, savings minus the operation and

maintenance costs; IR, interest rate; n, payback period; P, present worth capital costs

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