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ORIGINAL ARTICLE

Dual fluidized bed based technologies for carbon dioxide reduction — example hot metal production

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Abstract

The present work describes the results achieved during a study aiming at the full replacement of the natural gas demand of an integrated hot metal production. This work implements a novel approach using a biomass gasification plant combined with an electrolysis unit to substitute the present natural gas demand of an integrated hot metal production. Therefore, a simulation platform, including mathematical models for all relevant process units, enabling the calculation of all relevant mass and energy balances was created. As a result, the calculations show that a natural gas demand of about 385 MW can be replaced and an additional 100 MW hydrogen-rich reducing gas can be produced by the use of 132 MW of biomass together with 571 MW electricity produced from renewable energy. The results achieved indicate that a full replacement of the natural gas demand would be possible from a technological point of view. At the same time, the technological readiness level of available electrolysis units shows that a production at such a large scale has not been demonstrated yet.

Keywords Carbon dioxide reduction · Oxyfuel combustion · Sorption enhanced reforming · Biomass gasification

1 Introduction and short description

In the past, large amounts of easily accessible primary energy resources accompanied by an efficient energy infrastructure enabled the development of pleasant wealth in Europe. At the same time, limited resources in Europe itself led to significant dependency on energy imports. The energy strategy of the European Union for the future aims at pretending secure, safe and affordable energy. Furthermore, the energy strategy includes the utilization of local available resources, a reduction of greenhouse gas emissions, and the development of new innovative energy technologies, as new high performance low-carbon technologies [1, 2].

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The production of hot metal causes significant fossil carbon dioxide (CO_2) emissions. Therefore, numerous researchers investigate the reduction of fossil carbon dioxide emission in the surrounding of hot metal production. Hereby, the replacement of fossil energy carriers should not impair the quality of final products or lead to a reduced availability of the applied production process.

So far, the following question:

Which setup in an existing integrated hot metal production would enable the most reasonable reduction of fossil carbon usage based on available dual fluidized bed based technologies?

has not been answered. The following paper describes the results of investigations aiming at reasonable changes with respect to the reduction of fossil carbon usage of a hot metal production process. At the beginning, a short review about the potential application of dual fluidized bed based technologies is carried out [3]. Afterwards, an application for hot metal production is investigated. This step bases on the creation of an optimized industrial model of the integrated hot metal production. Within the present work, a description of the production process is carried out. Furthermore, the present paper discusses:

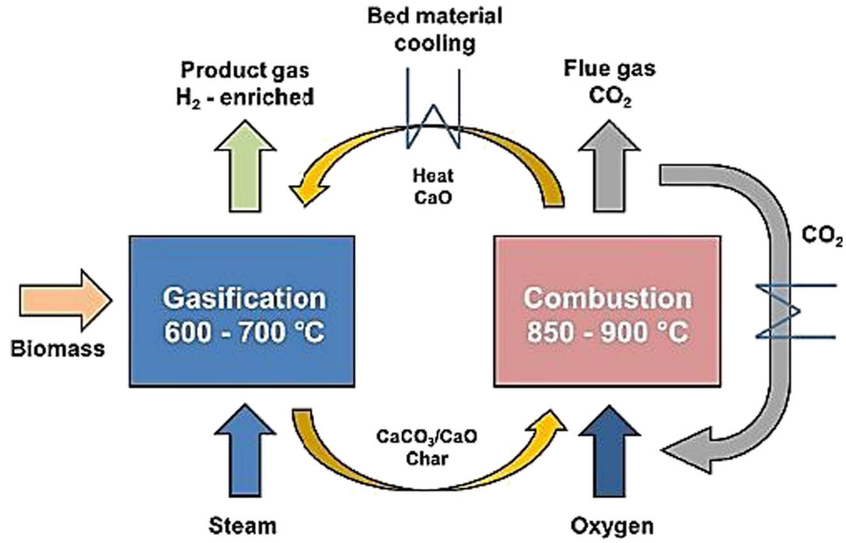

Fig. 1 Sorption enhanced reforming with oxyfuel combustion (OxySER)

- The application of dual fluidized bed technology for carbon dioxide reduction,
- The used concept for the reduction of fossil-based carbon emissions,
- The creation of an industrial plant model,
- The simulation results achieved,
- And an interpretation of the achieved results with respect to a large-scale implementation.

Herby, within the present work, an industrial plant information model is defined as a simplified reproduction of the reality of an industrial plant focused on the investigation of important existing relations. This includes beside technical parameters such as fundamental chemical and thermodynamic relations also practical aspects for the operation such as operation control, economic performance, ecologic impact and legal aspects relevant for an optimized future operation. Following this definition, the focus of the illustrated model within the present work lies on the mass- and energy balances of a large-scale plant with respect to an ecologic optimization by a reduction of the fossil carbon utilization.

2 Technological review

The dual fluidized bed (DFB) technology offers a broad range of applications for the utilization of CO_2 neutral energy carriers like biomass. Conventional biomass steam gasification is an already well-known technology. Based on this technology the sorption enhanced reforming process (SER) was developed and enables the in-situ removal of CO_2 from the product gas. Consequently, the chemical equilibrium of the product gas is shifted and a high hydrogen (H_2) content can be obtained in the product gas. By applying oxyfuel combustion in the combustion reactor (OxySER), an almost pure CO_2 stream can be obtained [3]. Thus, both gas streams can contribute to a CO_2

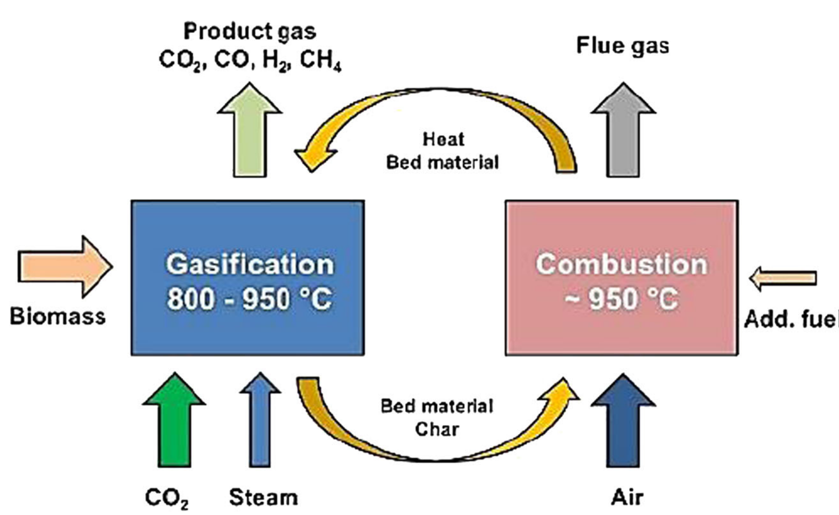

Fig. 2 CO_2 and steam gasification within a DFB reactor system

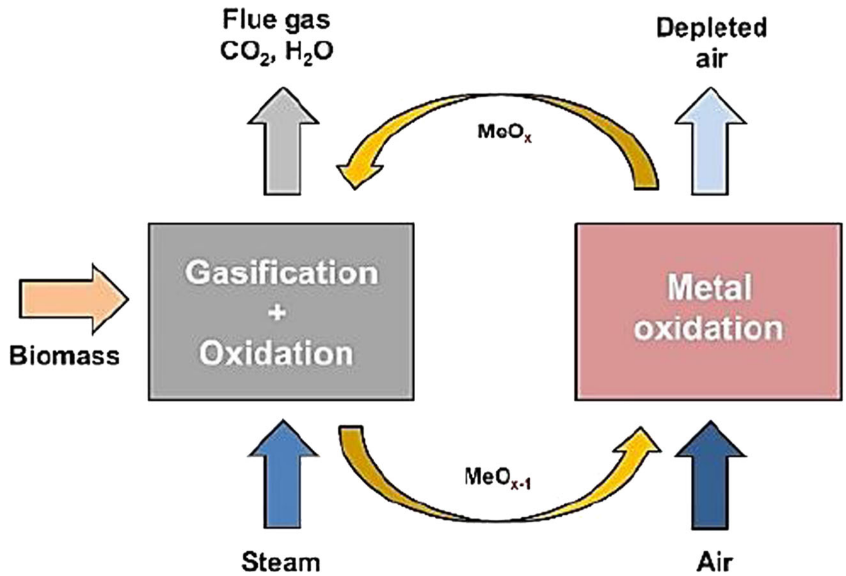

Fig. 3 Chemical looping combustion of solid biogenic fuels within a DFB reactor system

reduction in the atmosphere: On the one hand, a hydrogen-rich product gas is generated, which can be used directly as a reducing agent in steel industry or as basis for synthesis processes like methanation. On the other hand, a pure CO_2 stream is produced, which either can be stored or, again, used for synthesis processes like methanation [4]. A scheme of the process can be found in Fig. 1 [5].

Another variation of a DFB process is the gasification with CO_2 instead of steam shown in Fig. 2. The usage of CO_2 leads to a product gas with high carbon content. This seems to be contradictory, since decarbonization is the overall aim of energy intensive industries. Nevertheless, carbon, although it might be provided as CO_2 or CO respectively, is an important element for many processes and could be reused in this way to generate a product gas as basis for further utilization [6, 7].

Last but not least, a DFB reactor system can also be used for the so-called chemical looping combustion of solid biogenic fuels (BioCLC) process, which has promising potential for capturing CO_2 due to its low energy demand. The principle of the chemical looping process is shown in Fig. 3 and is based on the use of a metal oxide as bed material and oxygen carrier. This oxygen carrier is used to burn the gas, which is produced by the gasification reactions of the biomass with steam. The oxygen carrier itself is oxidized in the air reactor again. This procedure enables the production of a nitrogen-free flue gas. Therefore, the gas consists mainly of CO_2 and H_2O [8].

Several experimental campaigns have been conducted with the advanced 100 kW_{th} pilot plant at TU Wien to investigate the different technological approaches. The mentioned pilot plant consists of a gasification reactor and a combustion reactor with an overall height of about 7 m. More details about the latest reactor-design can be found in literature [9]. Figure 4 shows pictures of the described test plant.

The three very different dual fluidized bed processes offer different advantages and disadvantages. Table 1

Fig. 4 Upper part of the 100 kWth dual fluidized bed test plant (left), lower part (right) [10]

Table 1 Comparison of presented processes regarding different parameters

	OxySER [11, 12]	CO_2 and steam gasification of biomass [13, 14]	BioCLC [15, 16]
Achieved products	Hydrogen-rich product gas*, CO_2 and heat	Carbon monoxide-rich product gas* and heat	Carbon dioxide and heat*
Technical readiness	Pilot scale	Pilot scale	Pilot scale
Advantages	Product gas with high H_2 content and pure CO_2 for further utilization	Utilization of CO_2 as gasification agent enabling the reuse of carbon.	Pure CO_2 from biomass for further utilization
Disadvantages	Air separation unit necessary for oxygen supply	limited CO_2 conversion efficiency	No chemical energy supply
CO_2-avoidance (t/MWh_{out})*	~0.35	0.25–0.50	0.25–0.50
CO_2-capture (t/MWh_{out})*	~0.25	-	~0.4
* Reference	Investigated case in present work	Synthesis gas from coal	Conventional combustion process

provides and overview about different parameters of these processes. Typically, the gasification processes OxySER and CO_2 gasification produce a product gas, which contains chemical energy, whereas the CLC process as a typical combustion process provides heat. Especially, the OxySER process offers advantages, but also disadvantages. On the one hand, a hydrogen-rich product gas is produced; on the other hand, carbon from biomass is selectively transported to a separate CO_2 stream. Since biomass is the only renewable carbon source, the OxySER concept must be implemented wisely. This means the integration in an environment where H_2 on the one hand, but pure CO_2 on the other hand is needed. Therefore, within the present work, the application of OxySER in the surrounding of hot metal production is investigated.

3 Methodology for application in hot metal production

Figure 5 shows the used concept for the creation of an industrial plant model enabling a full replacement of the natural gas supply of a hot metal production. As can be seen, the proposed concept consists of a biomass gasification system, an electrolysis unit, a carbon dioxide removal unit and a methanation unit. The used biomass gasification system is operated as dual fluidized bed gasification system converting woody biomass into a hydrogen-rich gas. Besides, the used operation mode enables the production of a carbon dioxide-rich gas to follow a carbon capture perspective and enable further utilization. Figure 6 gives a detailed illustration of the basic principle of this process, which has been investigated intensively in the

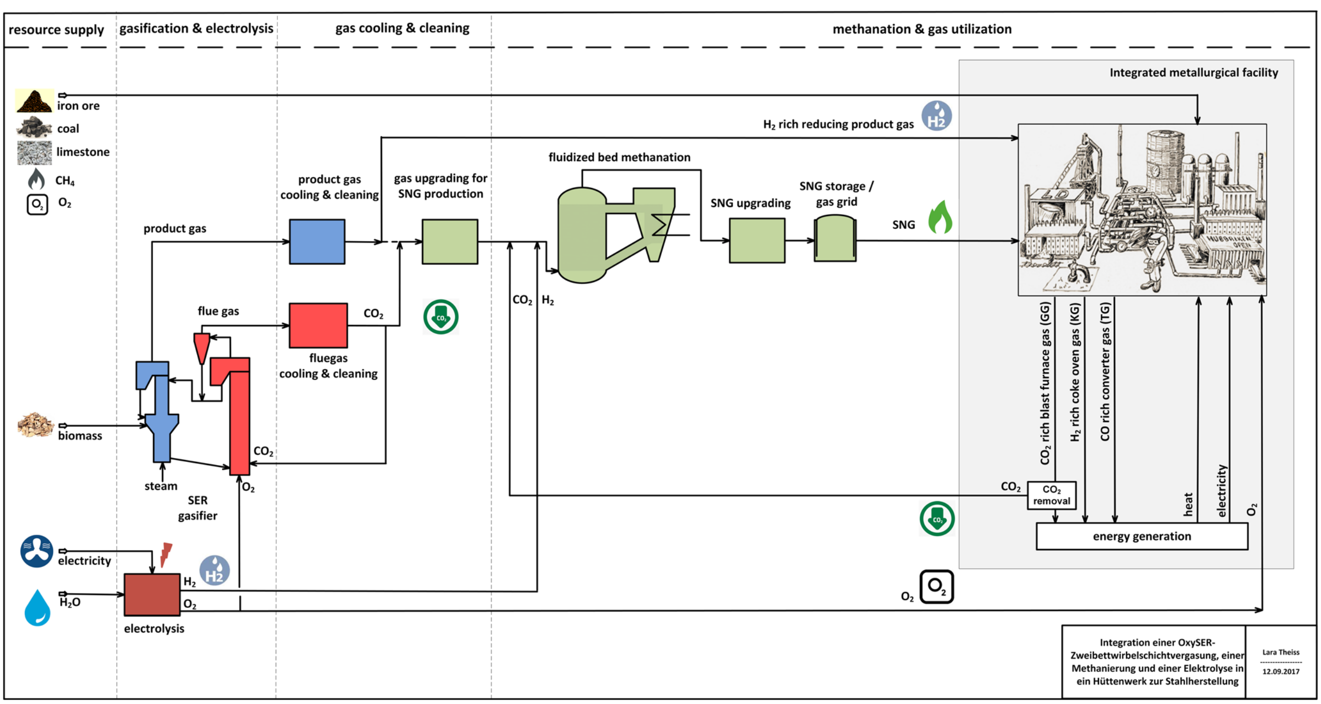

Fig. 5 Concept for full replacement of the natural gas demand of a hot metal production

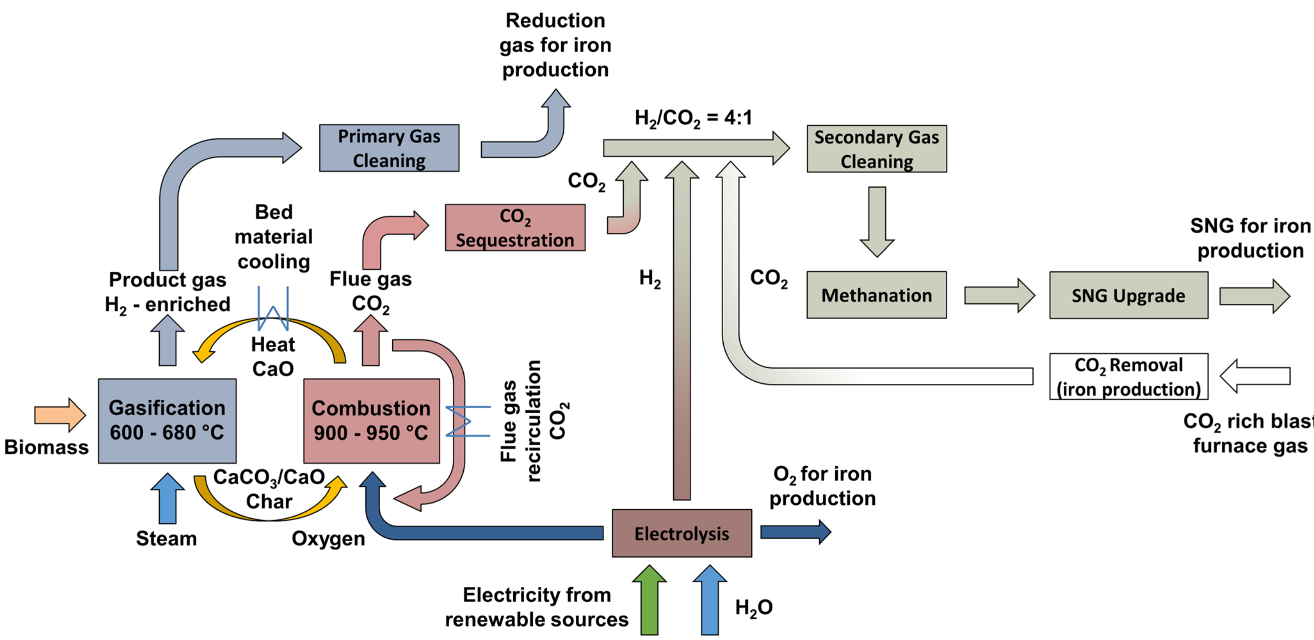

Fig. 6 Schematic illustration of sorption enhanced reforming in combination with oxyfuel combustion (OxySER), subsequent gas cleaning, electrolysis, and CO_2 separation

last years [17–19]. A detailed description of the so-called OxySER process can be found in literature [20, 27, 28].

Furthermore, the described concept in Fig. 5 includes an electrolysis unit. The electrolysis unit is used to provide

Table 2 Comparison of main important gas streams within the investigated process model

Parameter	Unit	Coke oven gas	Converter gas	Blast furnace gas	Product gas OxySER [19]	Flue gas OxySER [20]	Requirements methanation [26]
Hydrogen (H_2)	vol.-%$_{db}$	66	0.6	3.7	70	-	30–90
Carbon monoxide (CO)	vol.-%$_{db}$	5.8	51.8	25	8	-	0–25
Carbon dioxide (CO_2)	vol.-%$_{db}$	1.2	20	23	8	91	0–30
Methane (CH_4)	vol.-%$_{db}$	22	-	-	11	-	0–100
Nitrogen (N_2)	vol.-%$_{db}$	3	27.6	48.3	-	-	< 3
Non condensable C_xH_y	vol.-%$_{db}$	2	-	-	3	-	x
Oxygen (O_2)	vol.-%$_{db}$	-	-	-	-	9	x
Dust particles	mg/Nm^3	1.3	0.8	0.6	30	x	< 0.5
Sulfur: H_2S, COS, CS_2	mg/Nm$^3_{db}$	250	-	208	122	-	< 0.4
Nitrogen: NH3, HCN	mg/Nm$^3_{db}$	510	-	0.4	13	-	< 0.8
Halogens: HCl, HBr, HF	mg/Nm$^3_{db}$	-	-	5.8	12	-	< 0.06
Alkali metals: K, Na	mg/Nm$^3_{db}$	-	-	-	x	-	< 1
Tar	mg/Nm$^3_{db}$	x	x	x	25	-	< 0.1
Nitrogen oxide as NO_2	mg/Nm$^3_{db}$	x	x	x	-	900	x
Sulfur dioxide (SO_2)	mg/Nm$^3_{db}$	x	x	11.9	-	< 3	x
Volume flow	Nm^3/h	75,000	50,000	1,000,000			
Lower heating value	MJ/Nm$^3_{db}$	17.3	6.8	4.0	14.7		

x not available and experimental determination recommended

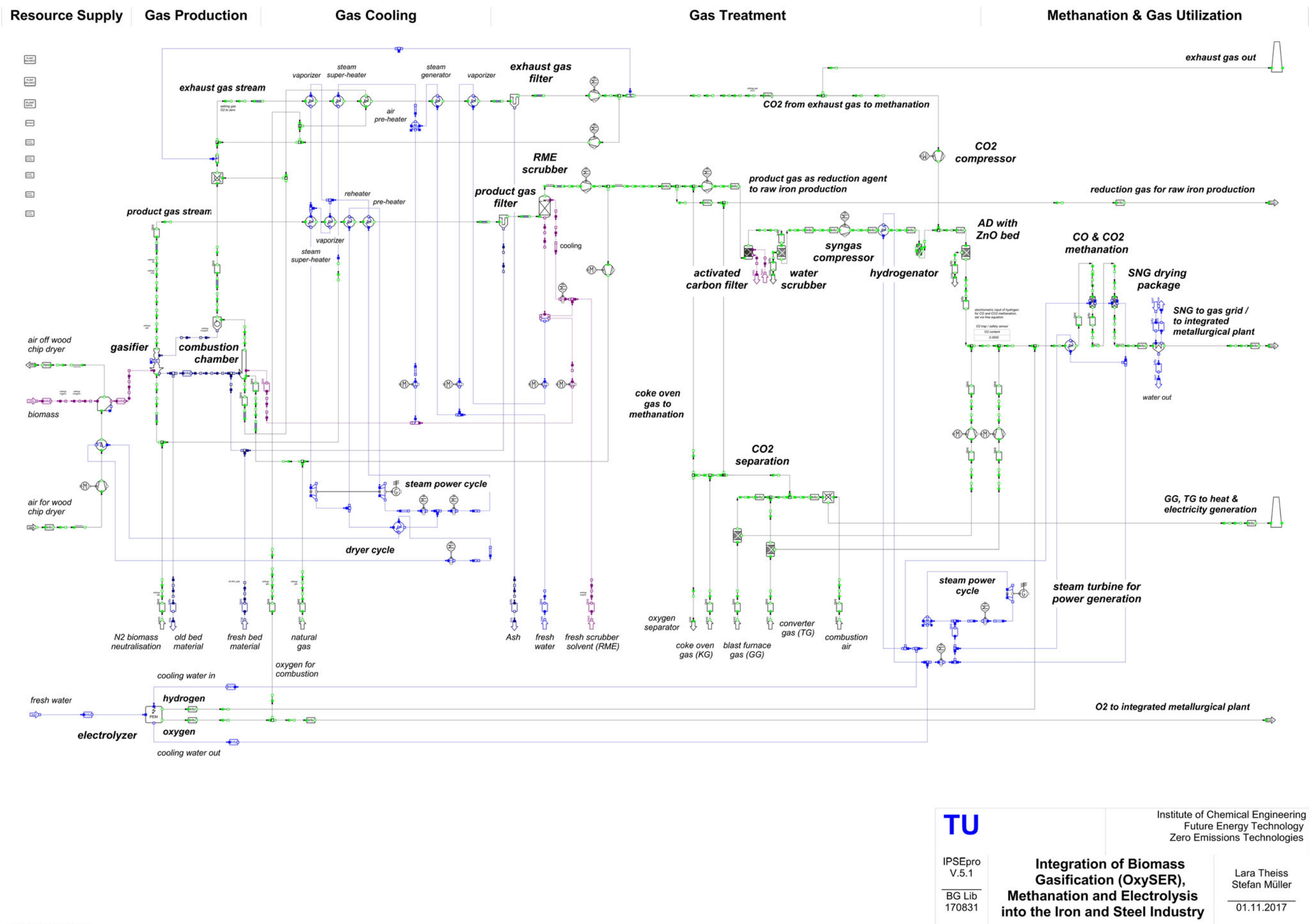

Fig. 7 Flowchart of the created process simulation model

hydrogen to the overall process allowing methanation according to stoichiometric ideal conditions accounting present biogenic carbon as well as recycled carbon dioxide from a blast furnace. A carbon dioxide removal unit is used to recycle carbon dioxide from the CO_2-rich blast furnace gas as a feedstock for the methanation process. A detailed description of the process units can be found elsewhere [21–23]. Figure 6 shows a simplified sketch of the process concept.

Hereby, the applied biomass gasification system and the electrolysis are used to replace most parts of the fossil energy demand of a hot metal production [24]. To achieve this, the electricity demand for electrolysis is delivered by renewable sources like wind, water or sun power. As a result, a hydrogen-rich reducing gas as well as a synthetic natural gas (SNG) is supplied to the production process. At this point, it must be mentioned that the illustrated concept is not influenc-

Table 3 Results of the mass and energy balance calculations

Input parameter	Unit	Value	Output parameter	Unit	Value
Mass balance					
Wood chips dry	kg/h	37468*	H_2-rich product gas	kg/h	15,671
Carbon dioxide (CO_2)	kg/h	40,247	Synthetic natural gas	kg/h	28,206
Fresh water	kg/h	60,365	Oxygen (O_2)	kg/h	96,325
Others	kg/h	5791	Others	kg/h	3669
Energy utilization:					
Wood chips wet	MW	132	H_2-rich product gas	MW	100
Electricity electrolysis	MW	571	Synthetic natural gas	MW	385

*50 t/h before dryer

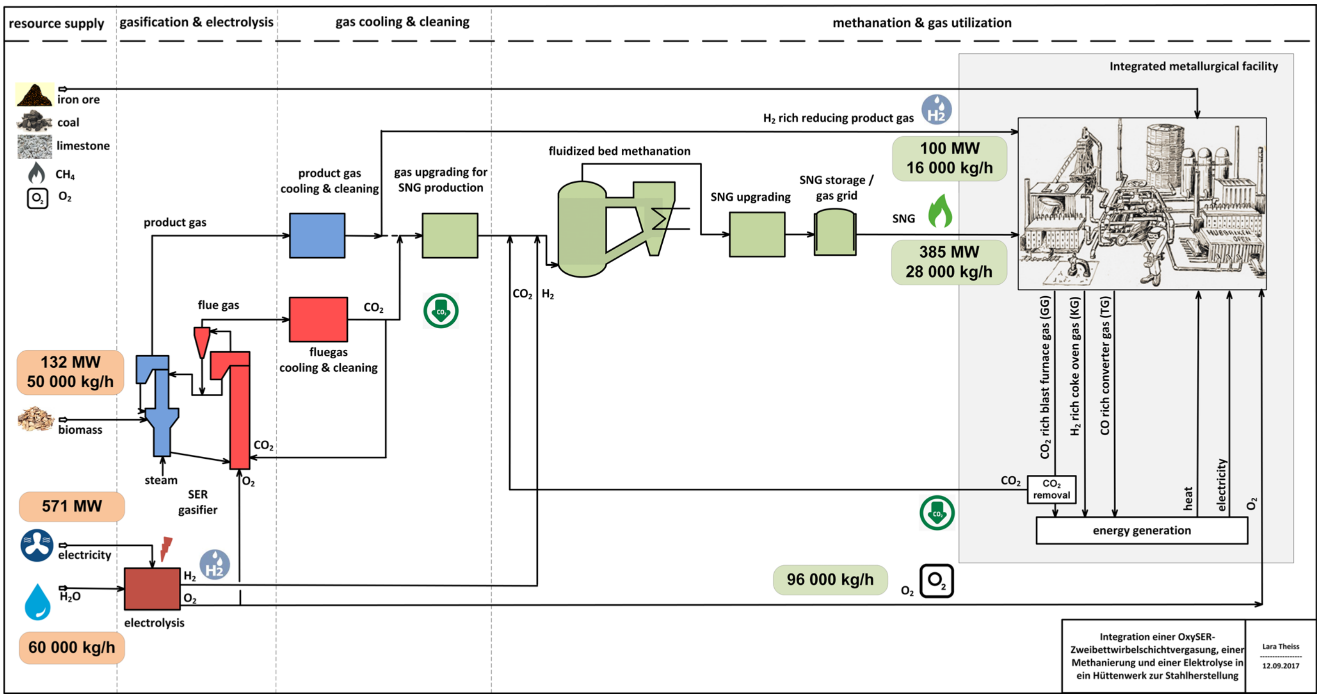

Fig. 8 Optimized industrial plant model with reduced fossil carbon usage

ing the existing core production process and therefore ensures the desired quality of final products. The described process concept has been used for the creation of a digital industrial plant information model (IPIM) [25] allowing the calculation of all relevant information with respect to the investigated process concept. The executed work included:

- A data analysis of existing mass and energy streams,
- Modeling of an optimized conceptual design (cf. Fig. 4),
- The creation of an industrial plant information model for the
- Calculation of simulation results,
- As well as an interpretation of results achieved.

4 Results and discussion

Table 2 gives an overview about main relevant data with respect to existing gas streams in an integrated steel mill. As can be seen, product gas from dual fluidized bed biomass gasification shows similar properties like coke oven gas. Therefore, the product gas represents a valuable gas for a direct utilization within an integrated steel mill. In comparison, the converter gas and blast furnace gas show significantly lower heating values. Therefore, this gas streams can be considered containing low value with respect to its utilization possibilities. So, this gas streams are only used for the generation of process heat. At the same time, the blast furnace gas represents the

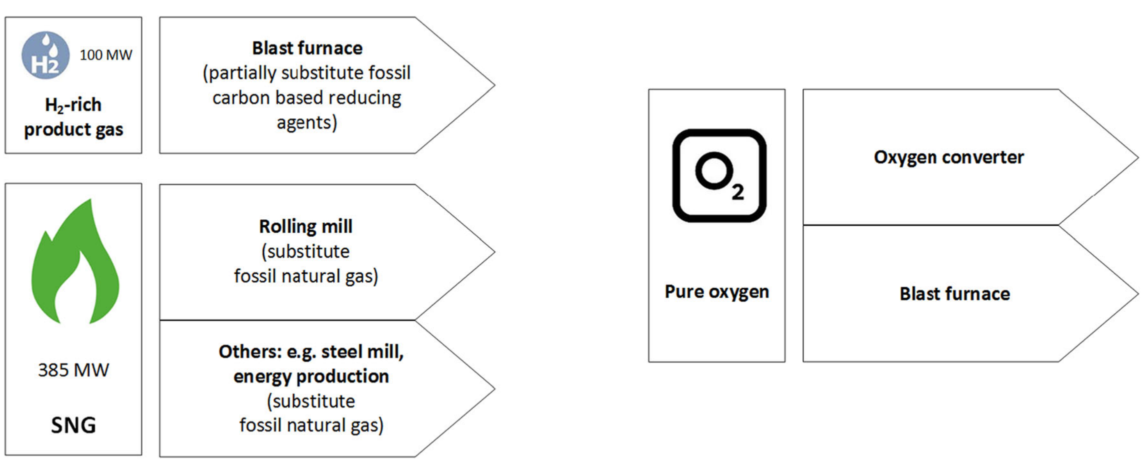

Fig. 9 Applications of the generated product streams

largest off-gas stream containing a high share of carbon dioxide. A process configuration, which is reusing the carbon dioxide from this stream, would enable a high carbon recycling rate and lead to significantly lower carbon dioxide emissions. The described circumstances formed the basis for the created concept displayed in Fig. 5.

Figure 7 shows a process flow diagram of the created process simulation model. It contains all relevant process units and parameters in the scale of an existing hot metal production. The used model library has been described by [29] with modifications and additions made by the authors of the present work [30]. The calculation of the OxySER process bases on experimental data from TU Wien and Stuttgart. Besides, it takes into account made experiences with the biomass gasification plants in Güssing, Oberwart, and Senden [25]. The calculation of the electrolysis was carried out based on provided data from literature [21]. The electrolysis unit supplies additional oxygen (O_2), which can be supplied to the hot metal production to cover internal demand. The calculation of the methanation of reused carbon dioxide with hydrogen produced by water electrolysis as well as the carbon dioxide from the OxySER has been carried out based on experiences provided by several authors from literature [31]. Described sources have been used for the creation of the simulation model.

Within the simulation model, available high temperature heat is used to produce electricity via a steam cycle process. The created simulation model was implemented into the surrounding of a hot metal production. As a result, Table 3 summarizes the main calculation results achieved and Fig. 8 shows a simplified illustration of main calculation results. 50 t/h of wood chips and 571 MW of electricity are necessary to enable a substitution of the present natural gas demand of 385 MW. The products obtained by the OxySER process, methanation, and excess oxygen from electrolysis as can be seen in Fig. 9 can be fully used within the hot metal production plant:

i. The OxySER product gas serves as reducing agent within the blast furnace and thereby replaces fossil pulverized coal injection (PCI) coal.
ii. The produced SNG replaces the total natural gas demand of the hot metal production. Natural gas is mainly used in the rolling mill, steel mill and for energy production in an integrated hot metal production.
iii. The excess oxygen from electrolysis is used in integrated metallurgical facility for example in an oxygen converter and the blast furnace.

The process corresponds to a possible equivalent CO_2 reduction of 77 t/h due to the replacement of fossil natural gas. Additionally, 100 MW of a valuable hydrogen-rich product gas can be supplied to the hot metal reduction process, which could reduce equivalent fossil CO_2 emissions by 33 t/h due to the replacement of PCI coal [32]. 40 t/h of carbon dioxide needs to be recycled from the blast furnace gas to supply enough carbon for the methanation unit. The implemented reuse of carbon dioxide would represent an interesting approach for the recycling of fossil carbon following the idea of a circular economy action plan [33]. As by-product from the electrolysis, 96 t/h of pure oxygen are produced. Illustrated numbers indicate the expected size of the proposed process concept if a full replacement of the present natural gas demand is the aim of further implementation steps.

5 Conclusion and outlook

The present work was executed to determine possible modifications to enable a significant reduction of fossil carbon dioxide emissions of a hot metal production. Biomass gasification, electrolysis, and carbon dioxide separation was identified as possible short-term modifications to enable a full replacement of the present natural gas demand. The proposed integration of the identified process units was calculated by the use of a process simulation model. As a result, the proposed modifications are not expected to cause negative influence on the quality of final products. As a further result, the carried out calculations show that:

- 50 t/h of woody biomass,
- 60 t/h of water,
- And 571 MW of electricity

would be necessary to replace fossil natural gas. This would represent about 1% of the primary energy usage of woody biomass in the energy sector in Austria [34]. Besides, the recycling of 20,000 Nm^3/h of carbon dioxide (CO_2) is required to operate the proposed process configuration. This would represent a reuse of consumed fossil and biogenic carbon sources. The investigated process within the present work could enable a saving of about 800,000 tCO_2/a. An estimation indicates that this represents a share of up to 1% of the overall CO_2 emissions in Austria [35]. The achieved results show that the natural gas demand of an integrated hot metal production can be replaced by the use of electrolysis, biomass gasification and a methanation system. The created model indicates valuable data for the design of proposed modifications. The following steps:

- Up-scaling of OxySER to 150 MW,
- Up-scaling of electrolysis to 600 MW,

and long-term tests of the methanation step with real gas from hot metal production are recommended to be executed before an implementation at a larger scale. These steps are necessary, as so far, no electrolysis unit at this scale is available, whereas, the largest dual fluidized bed biomass gasification system so far has been built in the scale of 32 MW in Gothenburg, Sweden.

Furthermore, a detailed analysis of the economic circumstances is recommended in near future. Recently published results indicate important economic aspects with respect to a further implementation of the presented concept. Main findings show that biomass based concepts could be economically feasible if the biomass price, the natural gas price and the price for CO_2 emission certificates provide a reasonable development with respect to the political aims formulated by the European Commission [36, 37]. A quick acceleration of accompanying implementation steps is demanded, if there should be any chance to reach the climate targets from the latest Paris Agreement.

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Abbreviations BioCLC, chemical looping combustion of solid biogenic fuels; CO_2, carbon dioxide; DFB, dual fluidized bed; DME, dimethyl ether; H_2, hydrogen; IPIM, industrial plant information model; O_2, oxygen; OxySER, sorption enhanced reforming in combination with oxyfuel combustion; PCI, pulverized coal injection; SER, sorption enhanced reforming; SNG, synthetic natural gas

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