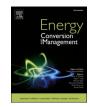


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# Full oxygen blast furnace steelmaking: From direct hydrogen injection to methanized BFG injection



# Jorge Perpiñán<sup>\*</sup>, Manuel Bailera, Begoña Peña

Escuela de Ingeniería y Arquitectura, Universidad de Zaragoza, María de Luna 3, 50018 Zaragoza, Spain

#### ARTICLE INFO

#### ABSTRACT

Keywords: Iron and steel industry Power to Gas Methanation Hydrogen injection Decarbonization Electrification This paper presents a novel concept of Power to Gas in an oxygen blast furnace, through blast furnace gas methanation and direct  $H_2$  injection. The PEM electrolyser produces  $H_2$ , which reacts with the CO and CO<sub>2</sub> from the blast furnace gas forming synthetic natural gas. The latter gas is injected into the blast furnace, closing a carbon loop and avoiding CO<sub>2</sub> emissions. A parametric analysis is performed to vary the  $H_2$ :CO<sub>2</sub> ratio in the methanation reaction. Different ratios are simulated and compared, among of which the most representative are: (i) 2.5, where unreacted CO<sub>2</sub> is directly recycled with the synthetic natural gas; (ii) 4, where stoichiometric conditions are found and the synthetic gas is composed mostly by CH<sub>4</sub>; and (iii) 8, where an excess of  $H_2$  is found in the synthetic gas; and (iv) an infinite ratio, where only  $H_2$  is injected in the blast furnace. In the latter, the methanation plant is not required, and no synthetic natural gas is produced. The results show that low  $H_2$ :CO<sub>2</sub> ratios perform poorly, involving high PEM sizes and high costs but only a 5% of CO<sub>2</sub> avoidance (compared to conventional blast furnaces). A  $H_2$ :CO<sub>2</sub> ratio of 4 and full  $H_2$  injection results in higher reduction of CO<sub>2</sub> emissions (33.8 % and 28.6%) with carbon abatement costs of 260 and 245  $\epsilon/t_{CO2}$ , respectively.

# 1. Introduction

The iron and steel (I&S) industry is one of the main contributors to global  $CO_2$  emissions with up to 27% of the global manufacturing sector [1]. Over 70 % of these emissions are produced by ironmaking blast furnaces (BF), which also consume the largest amount of energy in the entire industry (13–14 GJ per ton of hot metal, t<sub>HM</sub> in the following) [1].

The dominant method of steel production worldwide is the blast furnace-basic oxygen furnace (BF-BOF) route, which accounts for over 70% of global output. This process involves the preparation of raw materials including iron ore, coke, and limestone. Iron ore is agglomerated through sintering, coke is produced from coal, and limestone is calcined to obtain lime [2,3]. These materials are then fed into the blast furnace (BF), where coke supplies heat and CO-reducing gas that reacts with iron oxides [4]. The resulting molten iron is further refined in the basic oxygen furnace (BOF) by injecting pure oxygen to reduce carbon content. Finally, the refined steel is cast into various shapes, cooled, and after that undergoes finishing techniques for its desired applications.

Given the current policies of the European Union focused on the ambitious target of zero emissions by 2050 [5], I&S industry is very interested in the development of low-carbon technologies [6–8]. Various

strategies are being explored for  $CO_2$  abatement in blast furnace ironmaking. These strategies include injecting low-carbon reducing agents like hydrogen, implementing Top Gas Recycling (TGR) in oxygen blast furnaces (OBF), employing Carbon Capture and Storage (CCS), and applying Power to X (PtX) technologies like Power to Methane. These approaches aim to reduce greenhouse gas emissions and contribute to the decarbonisation of the ironmaking industry.

Most efforts have focused on the development of alternatives to coke, such as pulverized coal injection, natural gas, coke oven gas, waste plastics or other hydrocarbon injections [4], and more also synthetic natural gas, bio-methane, syngas from biomass or hydrogen [9]. TGR has generated more attention over the past decades to achieve large  $CO_2$  emissions reductions, usually applied in oxygen blast furnaces (OBF), such as in the pilot plant (TRL 6–7) developed in the ULCOS project in Luleå [10]. This not only reduces emissions (by up to 40–55% [11]) but also increases energy efficiency and can lead to cost savings.

In OBFs the absence of nitrogen decreases the total bosh gas volume, requiring a preheating gas injection in the upper part of the furnace to maintain the required operation temperature. This injection, usually burnt blast furnace gas (BFG) at 1000 °C [12,13], also increases flexibility, as other reducing agents, such as natural gas injection or H<sub>2</sub>, may be used [12]. Additionally, the OBFs yield higher productivity rates than

\* Corresponding author. E-mail addresses: jorge.perpinan@unizar.es (J. Perpiñán), mbailera@unizar.es (M. Bailera), bpp@unizar.es (B. Peña).

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Nomenclature		HM	Hot Metal	
		I&S	Iron and Steel	
Abbrevi	ation Meaning	IEA	International Energy Agency	
AFT	Adiabatic Flame Temperature	LHV	Lower Heating Value	
ASU	Air Separation Unit	MDEA	Methyldiethanolamine	
AHF	Air Heating Furnace	PEM	Proton Exchange Membrane	
BF-BOF	Blast Furnace-Basic Oxygen Furnace	PtG	Power to Gas	
BFG	Blast Furnace Gas	PCI	Pulverised Coal Injection	
BOFG	Basic Oxygen Furnace Gas	PFD	Process Flow Diagram	
CC	Carbon Capture	SNG	Synthetic Natural Gas	
CCUS	Carbon Capture Utilization and Storage	t <sub>HM</sub>	Ton of Hot Metal	
COG	Coke Oven Gas	t <sub>CS</sub>	Ton of Crude Steel	
DRI	Direct Reduced Iron	TGR	Top Gas Recycling	
EAF	Electric Arc Furnace	TRL	Technology Readiness Level	
GHG	Green House Gas			

traditional BFs, by 30–100% according to different estimates [11]. One of the best options within the TGR technology is to combine it with vacuum pressure swing adsorption (VPSA) and oxygen blast furnace (TGR-OBF, TRL 5–6). In this process, the CO<sub>2</sub> is captured from the top gas (or from the BFG), while the remaining gases, mainly CO and H<sub>2</sub>, are recycled into the furnace [14–16]. CO<sub>2</sub> emissions of the blast furnace can be reduced between 26 % and 56 % with respect to conventional BFs [17], with capture costs ranging from 50  $\notin$ /t<sub>CO2</sub> to 90  $\notin$ /t<sub>CO2</sub> [18]. However, TGR-OBF concepts lead to a strong decrease in the available energy content for downstream processes, as they focus on CO recycling [19], being one of the main challenges presented for this kind of configurations.

Another potential low-carbon alternative is to apply carbon capture and storage (CCS) to conventional steelmaking plants (BF-BOF route). Among the available carbon capture technologies, the most promising are chemical absorption and physical adsorption. Both technologies have high TRL when applied in the I&S industry (TRL 6), being able to capture high amounts of  $CO_2$  (up to 1700 kg<sub>CO2</sub>/t<sub>HM</sub>) with reasonable costs (38–97 \$/t<sub>CO2</sub>) [18]. Other emerging carbon capture technologies that can be applied in the I&S industry are membranes and sorption enhanced water gas shift (SEWGS). Both technologies exhibit a low electricity penalty (1.2-2.4 MJe/kgCO2) and relatively low costs (35-60  $t_{CO2}$ , but right now only TRL 2–4 has been achieved [18]. The main limitations of CCS technology encompass various aspects, including the requirements for geological storage or large-scale and long-term carbon utilization, as well as the crucial factor of the public's acceptance, which impacts its widespread implementation [20]. Geological storage necessitates identifying suitable and secure underground reservoirs for permanent CO2 storage, ensuring their stability and integrity, and managing potential risks. Additionally, establishing the infrastructure and transportation systems for safe CO2 transport adds complexity and cost [21]. Concerns related to safety, environmental impact, and the potential for CO<sub>2</sub> leakage can shape public attitudes and influence the acceptance of CCS initiatives [22].

To integrate PtX in the I&S industry stands out as a promising alternative for reducing CO<sub>2</sub> emissions while providing additional benefits [9]. PtX technologies allow the indirect electrification of industrial processes through the generation of valuable products from renewable electricity, such as e-fuels or chemicals. PtX also allow direct electrification through the Power to Iron technology, which is investigated in the project ULCOLYSIS, but is still in the laboratory phase (TRL 2) [23]. When the intermediate energy carrier is present in gaseous form, usually H<sub>2</sub>, it is also called Power to Gas (PtG) [24]. The most common products of the PtG technology that can be applied in the steel industry are hydrogen (Power to Hydrogen) [25], methane (Power to Methane) [26,27] and syngas (Power to Syngas) [28]. Furthermore, hydrogen together with the top gas released from the BF can be utilized for

methanol synthesis, thereby producing a valuable raw material for the production of synthetic fuels, plastics, and basic chemicals [29].

Feasibility evaluations for Power to Hydrogen (PtH2) integration have primarily focused on air-blown blast furnaces, with only one study found on oxygen blast furnaces. Experimental testing of hydrogen injection has also been limited. Simulation studies using alkaline and solid oxide electrolyzers in air-blown blast furnaces have injected 20-33 kg/ tHM of preheated hydrogen at temperatures up to 1200 °C, resulting in CO2 emissions ranging from 1060 to 1182 kgCO2/tHM [25,27,30]. In an investigation by Bailera et al. [27], preheated hydrogen was injected into the tuyeres of an oxygen blast furnace with top gas recycling, using 24 kgH2/tHM and 321 kgTGR/tHM, leading to CO2 emissions of 905 kgCO2/tHM [27]. The study concluded that hydrogen injection was less polluting than synthetic natural gas injection but had a higher energy penalty due to the larger electrolyzer size. Thyssenkrupp [31] achieved the first hydrogen injection through one of the 28 tuyeres of blast furnace no. 9 in Duisburg in 2019, with plans to gradually expand the use of hydrogen to all tuyeres. The COURSE 50 project [32] conducted an experimental campaign from 2018 to 2020 using a 12 m3 experimental blast furnace, continuously injecting 32 kgH2/tHM and achieving a 16% reduction in CO2 emissions.

In the Power to Methane technology, methanation of carbonaceous compounds, such as CO and CO<sub>2</sub>, is achieved with H<sub>2</sub>, obtaining water as a by-product [33] through the Sabatier reaction. Few experimental studies were found regarding experimental methanation of steel gases (BFG, BOFG and COG) [34–37], reaching a TRL 4. Theoretical studies on the Power to Gas technology in the I&S industry have also been assessed, including configurations with biomass methanation [38], OBF [26,39], TGR and SOEC electrolysis [40]. These studies concluded that CO<sub>2</sub> avoidance ranging from 6% to 22% can be achieved, with energy penalties of approximately 18–34 MJ/kg<sub>CO2</sub>. The primary cost associated with this process is the electricity consumption from the electrolyser.

In previous works, the authors conducted comprehensive research in the field of iron and steel production. They developed and validated a benchmark model for the traditional I&S industry. Additionally, they explored the integration of Power to Gas with amine scrubbing in an airblown blast furnace, aiming to enhance its efficiency and reduce emissions [41]. Furthermore, the impact of oxygen enrichment and temperature on hot blast was analysed, providing valuable insights for improving the process [42]. The results of these studies revealed significant potential for CO<sub>2</sub> avoidance, with reductions ranging from 9% to 34% and related costs of  $52 - 233 \text{ €}/t_{\text{HM}}$  and  $283 - 352 \text{ €}/t_{\text{CO2}}$ .

The present work assesses the technical performance and cost of a PtG-steelmaking integration, where the blast furnace gas is directly diverted to a methanation stage. Combined with renewable  $H_2$  produced in a PEM electrolyser, the CO and CO<sub>2</sub> present in the BGF give rise to synthetic natural gas (SNG). Notably, this configuration eliminates the

need for a carbon capture stage since the blast furnace gas is directly methanized. The SNG is recycled back into the blast furnace, avoiding  $CO_2$  emissions and geological storage. This PtG configuration focuses on both  $CO_2$  and CO recycling via BFG methanation (contrary to the TGR configuration that only recycles CO), ensuring that all energy requirements of the downstream processes (sinter strand, power plant, etc.) are covered. This integration is studied by varying the H<sub>2</sub>:CO<sub>2</sub> ratio in the methanation stage, from a ratio of 2.5 (where part of the CO<sub>2</sub> is not methanized and injected in the furnace), to an infinite ratio (where only H<sub>2</sub> is found and injected in the furnace). The results are compared with a conventional I&S plant in terms of  $CO_2$  emissions, energy penalty and fossil fuel demand. Both simulations, the conventional plant and the PtG-steelmaking plant, have been modelled with Aspen Plus. An economic assessment of the PtG-steelmaking integration is made to identify the specific costs of the new integration.

# 2. Description of case studies

First, a conventional iron and steel (I&S) plant is considered as the reference case (Fig. 1, solid lines). The blast furnace (BF) is fed from the top with alternating layers of iron ore agglomerated in the sinter strand and coke produced in the corresponding oven. The hot metal results from the reduction process of iron in the BF with help of reducing agents, usually pulverized coal (PCI), which are injected from below together with the hot blast (air heated in the hot stoves). The hot metal (or pig iron) is mixed with scrap (recovered steel) and converted in crude steel after the excess of carbon is eliminated in the basic oxygen furnace (BOF). The required oxygen is provided by an air separation unit (ASU). The different process gases are used as fuels in different processes: blast furnace gas (BFG in cyan) in the sinter strand, hot stoves and power plant; basic oxygen furnace gas (BOFG, in brown) in the casting and power plant; and coke oven gas (COG in purple) in the coke oven, BOF and power plant. The integrated steelworks include a power plant to be electrically self-sufficient where the excess of process gases are burnt. The conventional I&S plant, which serves as a baseline for comparison in the present work, considers a 320  $t_{HM}$ /h blast furnace [43] and a 100 MW [44] power plant. The conventional I&S plant was previously modelled in Aspen Plus and validated with literature data [41].

The low-carbon concept consists of an oxygen blast furnace (OBF) with PtG integration, see Fig. 1, where the dashed lines represents the

changes regarding the base case. The main new blocks in this PtGsteelmaking integration are the PEM electrolyser for H<sub>2</sub> production (O<sub>2</sub> is also by-produced), and the methanation plant for synthetic natural gas (SNG) production (green lines). New streams (dashed lines) include pure oxygen in the OBF, some BFG as preheating gas injection, and some BFG diverted to the methanation. The two latter streams form a closed carbon loop, avoiding additional CO<sub>2</sub> emissions into the atmosphere. The oxygen from the PEM electrolyser is used to alleviate the ASU demand, feeding the OBF, the preheating gas injection and the BOF. The H<sub>2</sub>:CO<sub>2</sub> ratio of the methanation stage is gradually varied, from 2.5 mol<sub>H2</sub>/mol<sub>CO2</sub>, going through 4 mol<sub>H2</sub>/mol<sub>CO2</sub> (stoichiometric conditions), increasing the ratio to 8 mol<sub>H2</sub>/mol<sub>CO2</sub>, and reaching an infinite ratio (full H<sub>2</sub>, no BFG). In the last case, only H<sub>2</sub> is injected in the OBF, therefore no methanation stage is required and no SNG is produced.

# 3. Methodology

For the modelling and simulation of the conventional I&S plant (Section 3.1) and the PtG-steelmaking integration concepts (Section 3.2), the software Aspen Plus is used. Fig. 1 shows all hierarchy blocks included in the simulation. Each block has different sub-models and its own Aspen property method [45] in order to reproduce the process. In all cases, steady-state conditions, complete combustion and chemical equilibrium is assumed. The different concepts are compared with respect to the techno-economic indicators explained in Section 3.3.

# 3.1. Conventional I&S plant

The detailed description of the Aspen Plus model of the conventional I&S plant (solid lines in Fig. 1) can be found in [41]. The reference case encompasses multiple levels in different hierarchies, and different blocks are used to accurately simulate each process (see Supplementary data). BFG composition, flame temperature and air and coke flow rates are calculated through the Rist model [39,46]. The BFG composition is calculated as function of the utilization factors of CO and H<sub>2</sub> (Eq. (1) and Eq. (2), respectively). The CO (H<sub>2</sub>) utilization factor of a blast furnace refers to the ratio of CO (H<sub>2</sub>) used in the process to the total amount of CO and CO<sub>2</sub> (H<sub>2</sub> and H<sub>2</sub>O) gases produced. The power plant is a combined cycle with cooling between two air compression stages and three steam turbines. The ASU model is based in the cryogenic technology of

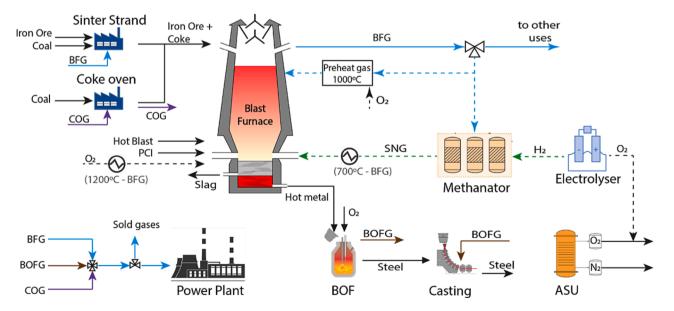


Fig. 1. Process flow diagram for conventional iron and steel plant (solid lines) and the integration of direct methanation through Power to Gas in an oxygen blast furnace (dashed lines). Blue line: blast furnace gas, BFG. Brown line: basic oxygen furnace gas, BOFG. Purple line: Coke oven gas, COG. Green line: synthetic gas, SNG.

Linde-Hampson model. The PENG-ROB property method is used for I&S processes and ASU, while PR-BM property method is used for the power plant.

$$\mu_{CO} = CO_2 / (CO_2 + CO) \tag{1}$$

$$\mu_{H2} = H_2 O / (H_2 O + H_2) \tag{2}$$

The model represents a simplified version of a real I&S plant, considering the design parameters summarized in Table 1 [41].

#### 3.2. PtG-steelmaking integration with direct BFG methanation

In the present work, the low-carbon I&S proposal integrates Power to Methane with an oxy blast furnace (OBF). The oxygen is injected in the OBF at 1200 °C, according to the better results obtained in our previous research [42] with respect to cold oxygen (25 °C) injection. As there is no nitrogen injected in the furnace, the insufficient preheat of the burden in the upper part of the furnace leads to BFG temperatures lower than 100 °C. To avoid problems related to oxidation and corrosion, a preheating gas injection in the upper part of the furnace is required. By doing so, the BFG temperature is maintained above 150 °C. The preheating gas consists of BFG burned with pure oxygen and injected at 1000 °C [12].

The distribution of the combustible gases remains the same as in the conventional I&S plant. The only differences are the absence of nitrogen in the BFG, resulting in lower mass flows, and that part of the BFG is now diverted to the preheating gas and the methanation plant (dashed blue lines in Fig. 1).

Regarding PtG technology, the PEM electrolyser is modelled with a specific energy consumption of 3.8 kWh/Nm<sup>3</sup>, working at ambient temperature. The methanation plant consists of two isothermal fixed-bed reactors, operating at 5 bar and 350 °C (first reactor) and 300 °C (second reactor), with an intermediate condensation stage at 100 °C (Fig. 2), in order to achieve a methane concentration in the SNG of 95% with stoichiometric conditions, see Eq. (3) and Eq. (4).

 $CO_2 + 4H_2 \leftrightarrow CH_4 + 2H_2O\Delta H_{298K} = -164.9kJ/mol$ (3)

$$CO + 3H_2 \leftrightarrow CH_4 + H_2O\Delta H_{298K} = -206.4kJ/mol \tag{4}$$

The PtG-steelmaking integration with direct BFG methanation is analysed under different operation conditions, obtained by varying the stoichiometry of the methanation reactor. Among others,  $H_2$ :CO<sub>2</sub> ratios of 2.5, 4 (stoichiometric conditions) and 8, to an infinite ratio (full  $H_2$ ) are simulated. When directly injecting  $H_2$ , no methanation stage is required, and therefore no SNG is produced. In all cases, the amount of SNG injected is limited to have an adiabatic flame temperature (AFT) of 2000 °C, because the larger the amount of SNG injected, the lower the

#### Table 1

Input data for the conventional I&S plant modelling [41].

Variable	Value	Units
Sintering temperature	1262	°C
Coke oven temperature	1100	°C
Hot blast pressure	4.5	bar
Hot blast temperature	1200	°C
Blast furnace top pressure	2.0	bar
H <sub>2</sub> utilization (Eq. (2))	0.47	-
Carbon content in hot metal	4.5	wt%
Carbon content in final steel	0.267	wt%
Basic oxygen furnace temperature	1650	°C
Scrap flow rate	150	kg/t <sub>HM</sub>
Separation pressure levels in ASU	6/1.5	bar
Gas turbine pressure (Power Plant)	27	bar
Gas turbine inlet temperature (Power Plant)	1389	°C
Steam inlet temperature (Power Plant)	560	°C
Steam pressure levels (Power Plant)	120/42/4	bar
Power plant size	100	MW

AFT is. If lower temperatures are found, productivity problems may arise [47]. The Power to Gas plant is sized according to the amount of SNG (or  $H_2$ ) that can be injected in the blast furnace. Thus, knowing the amount of synthetic gas required for each  $H_2$ :CO<sub>2</sub> ratio, the methanation plant and the electrolyser can be sized. The oxygen by-produced in the electrolyser is used to eliminate the ASU and to feed the BOF and hot blast enrichment necessities.

#### 3.3. Techno-economic indicators

The comparison of the proposed low-carbon concepts with the conventional I&S plant is performed with regards to different technoeconomic indicators.

The technical indicators are the coal-equivalent replacement ratio (coal-e RR) and the energy penalty. The former is defined as the sum of the coking coal and pulverized coal replaced by the SNG (Eq. (5)). The latter is the net energy consumed in the industry per kg of CO<sub>2</sub> avoided with the PtG-steelmaking integration (Eq. (6)). The involved magnitudes are detailed in Table 2.

$$Coal - eRR = \frac{\Delta \dot{m}_{PCI} + \Delta \dot{m}_{CokingCoal}}{\Delta \dot{m}_{SNG}} [kg_{Coal} / kg_{SNG}]$$
(5)

$$E_{penalty} = \frac{\Delta E_{cons} - \Delta E_{coal} \bullet \eta_{elec} - \Delta E_{gases} \bullet \eta_{elec}}{\Delta \dot{m}_{CO2}} [MJ/kg_{CO_2}]$$
(6)

The economic indicators are the specific costs per ton of  $CO_2$  avoided  $(\notin/t_{CO2})$  and per ton of hot metal  $(\notin/t_{HM})$ . The corresponding calculation is shown in Eq. (7) and Eq. (8), respectively, and the involved magnitudes are described in Table 2. The analysis assumed a loan amortization period of 20 years, a PtG system operating for 8000 h per year [48], and a renewable electricity price of 77  $\notin$ /MWh. The detailed calculation of CAPEX, OPEX and Incomes can be found elsewhere [41], but for completeness purposes it has been summarized in the supplementary data.

$$CO_{2}avoidanceCost = \frac{\left(\frac{Capex}{Loanamortization} + Opex - Incomes\right)\hat{A} \cdot 10^{6}}{CO_{2}avoided\hat{A} \cdot Operatinghours} \begin{bmatrix} \epsilon \\ t_{CO_{2}} \end{bmatrix}$$
(7)

$$SpecificImplementationCost = \frac{\left(\frac{Capex}{Loanamortization} + Opex - Incomes\right)\hat{A} \cdot 10^{6}}{IronProduction\hat{A} \cdot Operatinghours} \begin{bmatrix} \epsilon \\ I_{HRC} \end{bmatrix}$$
(8)

# 4. Results and discussion

Results and discussion on the PtG-steelmaking integration with oxy blast furnace and its comparison with the conventional I&S plant are presented in Sections 4.1 and 4.2 with respect to technical and economic aspects, respectively. The figures depict data presented as a function of the H<sub>2</sub>:CO<sub>2</sub> ratio in the methanation stage. The study includes a total of 12 data points, with the most significant ones being:  $2.5 \text{ mol}_{H2}/\text{mol}_{CO2}$ , 4 mol<sub>H2</sub>/mol<sub>CO2</sub> (stoichiometric conditions), and an infinite ratio (full H<sub>2</sub>).

## 4.1. PtG-steelmaking integration

In this section, we present the results of the integration of Power to Gas with steelmaking, specifically focusing on parameters like reducing agents, coal-e RR, SNG and BFG composition, PEM size,  $CO_2$  emissions and energy penalty.

Switching from air-blown BF (conventional I&S) to an OBF (PtGsteelmaking integration) enables higher amounts of reducing agents maintaining the AFT above 2000 °C. This configuration allows the full replacement of PCI by synthetic natural gas. When stoichiometric conditions are taken in the methanation plant (H<sub>2</sub>:CO<sub>2</sub> of 4), the minimum coke consumption in the OBF is obtained (280 kg/t<sub>HM</sub>), see Fig. 3,

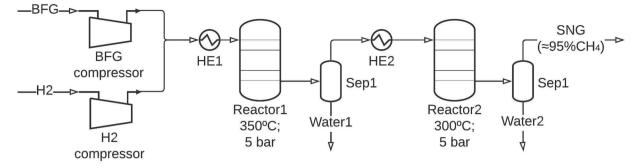


Fig. 2. Process flow diagram for the methanation plant (details in the text).

Table 2

Units and description	of variables fr	rom equations	(3)-(6).
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Variable	Units	Description
$\Delta \dot{m}_{PCI}$	$kg_{PCI}/t_{HM}$	Savings in PCI consumption
$\Delta \dot{m}_{CokingCoal}$	kg <sub>CokingCoal</sub> /t <sub>HM</sub>	Savings in coking coal consumption
$\Delta \dot{m}_{SNG}$	kg <sub>SNG</sub> /t <sub>HM</sub>	Increase in SNG injection
$\Delta E_{cons}$	$\mathrm{MJ}/\mathrm{t}_{\mathrm{HM}}$	Increase in electricity consumption in the industry
$\Delta E_{coal}$	$MJ/t_{HM}$	Savings in coal energy
$\Delta E_{gases}$	$MJ/t_{HM}$	Increase in excess sold gases
$\eta_{elec}$	-	Energy conversion factor from coal to electricity (0.33)
CAPEX	M€	Capital Expenditure
OPEX	M€/year	Operational Expenditure
Incomes	M€/year	Operational Incomes
Loan amortization	у	Process of paying off a loan over time
CO <sub>2</sub> avoided	t <sub>CO2</sub> /h	CO <sub>2</sub> avoided per hour
Operating hours	h/year	Operating hours per year

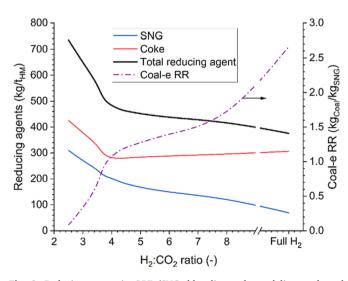


Fig. 3. Reducing agents in OBF (SNG, blue line; coke, red line; and total reducing agents, black line) and coal-e RR (purple dashed line) as a function of the  $H_2$ :CO<sub>2</sub> stoichiometry.

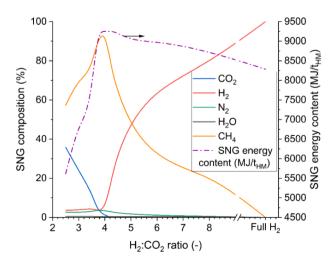
compared with that of the conventional I&S plant (332 kg<sub>coke</sub>/t<sub>HM</sub> and 150 kg<sub>PCI</sub>/t<sub>HM</sub>). When increasing the ratio towards full H<sub>2</sub> injection, 69 kg<sub>H2</sub>/t<sub>HM</sub> are injected, with a slight increase in the coke rate (307 kg/ t<sub>HM</sub>), but much less total reducing agent (376 kg/t<sub>HM</sub>) is needed. This is due to the higher coal-e RR of the H<sub>2</sub> when compared to the SNG. These results are consistent with previous studies conducted by Yilmaz et al. [25] and Kim et al. [30] in an air-blown blast furnace. Yilmaz injected hydrogen produced by an alkaline electrolyser, achieving a coke consumption of 390 kg<sub>coke</sub>/t<sub>HM</sub> and no use of PCI. Kim, on the other hand,

utilized hydrogen produced by a SOEC electrolyser, resulting in a coke consumption of 327 kg<sub>coke</sub>/t<sub>HM</sub> and a PCI consumption of 112 kg<sub>PCI</sub>/t<sub>HM</sub>. For H<sub>2</sub>:CO<sub>2</sub> ratios below 4, poor results are found. The coal-e RR drops below 1 and the coke rate increases considerably. This is because large amounts of CO<sub>2</sub> are not methanized and therefore are injected into the blast furnace.

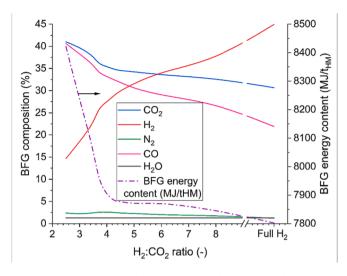
The BFG composition and the SNG composition are co-dependents of each other through the chemical reactions occurring in the blast furnace. The former gas is used in the methanation stage to form synthetic natural gas, which in turn is used as a reducing agent in the OBF, determining the BFG composition. The composition of both gases is also dependent on the H<sub>2</sub>:CO<sub>2</sub> ratio used in the methanation stage.

At a H<sub>2</sub>:CO<sub>2</sub> ratio of 4, the majority of the SNG consists of CH<sub>4</sub> (Fig. 4). As the ratio decreases, the CO<sub>2</sub> content increases, reaching 35.8 vol% at a ratio of 2.5, indicating direct recycling of CO<sub>2</sub>. On the other hand, higher ratios lead to an increase in H<sub>2</sub> content, up to 100% (full H<sub>2</sub>). The energy content of the SNG, shown by the purple dashed line in Fig. 4, takes into account both the lower heating value (LHV) of the gas (MJ/kg<sub>SNG</sub>) and the mass flow rate (kg<sub>SNG</sub>/t<sub>HM</sub>), providing the energy content of this stream in MJ/t<sub>HM</sub>. The maximum energy content of the SNG is observed at a H<sub>2</sub>:CO<sub>2</sub> ratio of 4. As the ratio decreases, more SNG is injected into the OBF, but a significant portion consists of CO<sub>2</sub>, resulting in lower energy content.

When  $CO_2$  is recycled through low  $H_2$ : $CO_2$  ratios, the BFG contains high amounts of CO and  $CO_2$  (Fig. 5). However, injecting pure  $H_2$  results in up to 45% hydrogen content in the gas. A stoichiometric  $H_2$ : $CO_2$  ratio results in 27%  $H_2$ , which is slightly higher compared to the 25%



**Fig. 4.** SNG composition ( $CO_2$ , blue line;  $H_2$ , red line;  $N_2$ , green line;  $H_2O$ , grey line;  $CH_4$ , orange line) and SNG energy content (purple dashed line) as a function of the  $H_2$ : $CO_2$  stoichiometry.

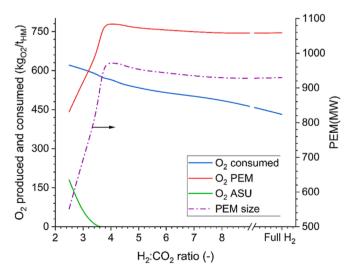


**Fig. 5.** BFG composition (CO<sub>2</sub>, blue line;  $H_2$ , red line;  $N_2$ , green line; CO, pink line;  $H_2O$ , grey line) and BFG energy content (purple dashed line) as a function of the  $H_2$ :CO<sub>2</sub> stoichiometry.

reported by Sato et al. [13] in their simulation of an OBF with high natural gas injection. Despite the increase in CO<sub>2</sub> and decrease in H<sub>2</sub>, a lower H<sub>2</sub>:CO<sub>2</sub> ratio leads to a higher BFG energy content of 8424 MJ/ t<sub>HM</sub>. This is due to the blast furnace's increased coke rate, resulting in higher amounts of CO and larger BFG mass flow rates. For a stoichiometric H<sub>2</sub>:CO<sub>2</sub> ratio and full H<sub>2</sub> injection, the BFG energy content ranges between 7800 and 7900 MJ/t<sub>HM</sub>. In comparison to the conventional I&S plant's BFG energy content of 4930 MJ/t<sub>HM</sub>, all cases show a significant increase. The CO utilization ratio (Eq. (1)) increases from 0.49 at a fixed H<sub>2</sub>:CO<sub>2</sub> ratio of 2.5 to 0.52 at the stoichiometric ratio and further to 0.58 with full H<sub>2</sub> injection.

A low H<sub>2</sub>:CO<sub>2</sub> ratio implies low H<sub>2</sub> demand, therefore the lowest PEM size is found at H<sub>2</sub>:CO<sub>2</sub> ratio of 2.5 (Fig. 6). At this point, the O<sub>2</sub> by-produced in the PEM is not able to supply the full oxygen demand. The deficit of oxygen is covered by the ASU. For a stoichiometric ratio and full H<sub>2</sub>, the PEM size ranges between 930 and 972 MW, being the lower size when pure hydrogen is injected in the OBF. The reason is that the H<sub>2</sub> cools the AFT to a larger degree, so less flow rate can be injected if compared with the SNG.

When the H<sub>2</sub>:CO<sub>2</sub> ratio is decreased, there is an increase in the



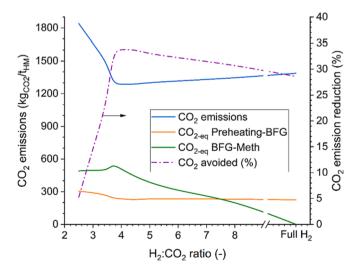
**Fig. 6.** Oxygen produced in the PEM (red line) and in the ASU (green line), oxygen consumed in the industry (blue line) and PEM capacity (purple dashed line) as a function of the  $H_2$ :CO<sub>2</sub> stoichiometry.

oxygen consumed by the industry (blue line) due to an increase in the oxy-hot blast mass flow rate (Fig. 6). When SNG is injected with  $CH_4$  as the primary component, additional oxygen is required in the OBF to partially oxidize methane into CO and  $H_2$ . However, only additional oxygen from the ASU is necessary at  $H_2$ :CO<sub>2</sub> ratios lower than 4.

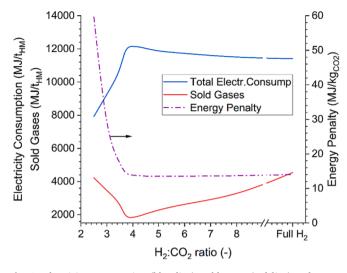
The integration of PtG with steelmaking shows that the optimum CO<sub>2</sub> emission reduction occurs at a stoichiometric H<sub>2</sub>:CO<sub>2</sub> ratio, reaching 33.8% of carbon avoidance (Fig. 7). However, as the H<sub>2</sub>:CO<sub>2</sub> ratio increases and full H<sub>2</sub> injection is employed, the percentage of CO<sub>2</sub> avoided decreases to 28.6%. This is primarily attributed to the coke rate, where higher carbon input in the form of coke leads to increased carbon output in the form of  $CO_2$  emissions. In Fig. 7, the orange and green lines represent the CO<sub>2-eq</sub> recycled within a closed loop through the preheating BFG gas and the BFG diverted to methanation, respectively. With full H<sub>2</sub> injection, no BFG is diverted to methanation, resulting in no recycling of CO<sub>2-eq</sub> through the PtG system. To provide context for these results, it is important to consider other low-carbon methods used in oxygen blast furnace ironmaking production, such as top gas recycling. Studies have shown that top gas recycling can achieve CO<sub>2</sub> emissions ranging from 868 to 1180  $t_{CO2}/t_{HM}$  in the blast furnace [27,49]. In comparison, the CO<sub>2</sub> emissions emitted in the blast furnace of PtG concepts of the present study range from 747 to 833  $t_{CO2}/t_{HM}$  (not to be confused with the total emissions of the entire I&S industry, which range from 1286 to 1387  $t_{CO2}/t_{HM}$ , shown in Fig. 7).

The sold gases, electricity consumption and energy penalty are shown in Fig. 8. The PEM electricity consumption accounts for 78–91% of the total electricity consumption, creating a linear dependency between the size of the electrolyser and the total electricity consumption. The maximum electricity consumption is 12163 MJ/t<sub>HM</sub> for a stoichiometric H<sub>2</sub>:CO<sub>2</sub> ratio, compared with the 950 MJ/t<sub>HM</sub> in the conventional I&S plant. When decreasing the H<sub>2</sub>:CO<sub>2</sub> ratio below 4, the ASU starts working, but its consumption is very low compared to that of the PEM electrolyser, barely affecting the results.

The amount of excess gases sold to nearby industries, including BFG, BOFG, and COG, decreases to 1828 MJ/t<sub>HM</sub> for a stoichiometric H<sub>2</sub>:CO<sub>2</sub> ratio (Fig. 8). However, for a H<sub>2</sub>:CO<sub>2</sub> ratio of 2.5 and full H<sub>2</sub> injection, the sold gases increase to over 4000 MJ/t<sub>HM</sub>. This indicates that the industry's energetic gases are better utilized when a H<sub>2</sub>:CO<sub>2</sub> ratio of 4 is employed. Lower H<sub>2</sub>:CO<sub>2</sub> ratios result in higher BFG energy content, while higher H<sub>2</sub>:CO<sub>2</sub> ratios lead to reduced BFG usage in the methanation process, resulting in larger quantities of gases available for sale.



**Fig. 7.**  $CO_2$  total emissions (blue line), CO2-eq in preheating BFG gas (orange line), CO2-eq in BFG diverted to methanation (green line) and percentage of  $CO_2$  emission reduction (purple dashed line) as a function of the H<sub>2</sub>:CO<sub>2</sub> stoichiometry. Orange and green lines represent the CO2-eq that is recycled in a closed loop.



**Fig. 8.** Electricity consumption (blue line), sold gases (red line) and energy penalty (purple dashed line) as a function of the H<sub>2</sub>:CO<sub>2</sub> stoichiometry.

The energy penalty increases drastically for lower  $H_2$ :CO<sub>2</sub> ratios because only small amounts of CO<sub>2</sub> are avoided (Fig. 8). For stoichiometric ratio and full  $H_2$  injection, the energy penalty is constant at 14 MJ/kg<sub>CO2</sub>. For a  $H_2$ :CO<sub>2</sub> ratio of 4, higher PEM sizes and electricity consumptions are found, but also higher amounts of avoided CO<sub>2</sub>, as opposed to full  $H_2$  injection, leading to a constant energy penalty.

For illustration purposes, two configurations analysed in this paper, namely stoichiometric  $H_2$ :CO<sub>2</sub> ratio and full  $H_2$  injection, are compared with OBF-TGR technology [27] (refer to Table 3). It's evident that the TGR configuration consumes 42–55% more fossil fuel (coke and PCI) than the other two configurations discussed. In terms of emissions from the blast furnace (BF) alone, the TGR setup exhibits higher CO<sub>2</sub> flow rates, but a significant portion can potentially be stored geologically. This implies that the environmental impact of the TGR configuration depends on the availability of storage. Furthermore, it's important to note that TGR technology often leads to a reduction in downstream process energy content, a drawback not observed in the other configurations explored here.

# 4.2. Economic analysis

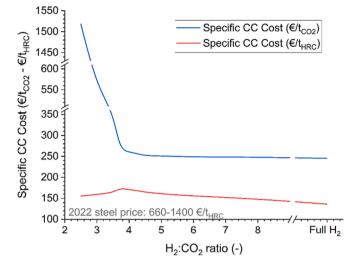
In this section, we present the economic results of integrating Power to Gas with steelmaking, focusing on parameters such as CAPEX, OPEX, Incomes and specific carbon avoidance costs.

Full H<sub>2</sub> injection yields the best economic results, with a cost of 245  $\notin/t_{CO2}$  and 136  $\notin/t_{HRC}$  (Fig. 9), despite having a lower CO<sub>2</sub> capture rate. This configuration has better results than a H<sub>2</sub>:CO<sub>2</sub> ratio of 4, which has a larger PEM size and costs 260  $\notin/t_{CO2}$  and 171  $\notin/t_{HRC}$ . The CAPEX, OPEX, and Incomes for both full H<sub>2</sub> injection and stoichiometric conditions are very similar, ranging from 870 to 931 M $\notin$ , 594–640 M%/year,

 Table 3

 Comparison of analysed configurations in this paper and TGR [27].

Variable	Units	Stoichiometric H <sub>2</sub> : CO <sub>2</sub> ratio	Full H <sub>2</sub> injection	TGR in oxy-BF
Coke	kg/t <sub>HM</sub>	280	307	235
PCI	kg/t <sub>HM</sub>	0	0	200
Gaseous reducing agent (SNG, H <sub>2</sub> or TGR)	kg/t <sub>HM</sub>	202	69	449
CO <sub>2</sub> emitted in BF	kg <sub>CO2</sub> ∕ t <sub>HM</sub>	747	833	1160
CO <sub>2</sub> to geological storage	kg <sub>CO2</sub> / t <sub>HM</sub>	0	0	500

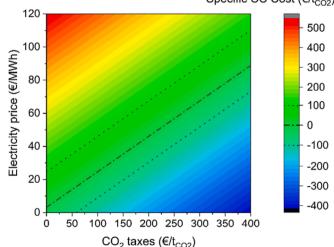


**Fig. 9.** Specific carbon capture (CC) cost in  $\ell$ /tCO<sub>2</sub> (blue line) and  $\ell$ /tHRC (red line) as a function of the H<sub>2</sub>:CO<sub>2</sub> stoichiometry (2022 steel price [50]).

and 248–288 M€/year, respectively. A H<sub>2</sub>:CO<sub>2</sub> ratio of 2.5 shows lower values for these parameters, resulting in a cost of 155 €/t<sub>HRC</sub>. However, due to poor CO<sub>2</sub> reduction, the specific carbon capture cost increases drastically to 1518 €/t<sub>CO2</sub>.

In all cases, the PEM contributes the most to the CAPEX, accounting for 35–43% of the total cost. Purchased electricity is the largest contributor to the OPEX, comprising 94–95% of the expenses. The primary source of income comes from CO<sub>2</sub> taxes, ranging from 33 to 57%. It is worth noting that the steel price in 2022 was around 660–1400  $€/t_{HRC}$ [50], so the specific carbon capture cost represents a 10–26% increase in the actual steel price, depending on the H<sub>2</sub>:CO<sub>2</sub> ratio. However, a recent study conducted by Subraveti et al. [51], published in January 2023, analysed CCS costs in a cement plant (90% capture with a 60% increase in cost) and a steel plant (47% capture with a 13% increase in cost), concluding that the cost increase in a bridge as the final product was only 1%. This implies that the costs incurred for implementing the PtG system in steelmaking, despite being significant, would have a limited impact on the final product competitiveness.

For the operation point with a stoichiometric H<sub>2</sub>:CO<sub>2</sub> ratio of 4, a sensitivity analysis focusing on the specific CC cost in  $\epsilon/t_{CO2}$  is performed (Fig. 10). In order to find the appropriate combinations of



Specific CC Cost (€/t<sub>CO2</sub>)

**Fig. 10.** Sensitivity analysis for stoichiometric ratio  $H_2$ :CO<sub>2</sub> of 4:1, representing specific CC cost in  $\ell/t_{CO2}$  as a function of the CO<sub>2</sub> taxes and the electricity price.

electricity price and CO<sub>2</sub> tax that lead to a profitable scenario, these two parameters are analysed between 0 and 120 €/MWh and 0–400 €/t<sub>CO2</sub>. With the current prices, the PtG-steelmaking integration is not profitable. However, under certain conditions negative values, e.g. actual benefits, can be obtained. For instance, 22 €/MWh or 345 €/t<sub>CO2</sub> is required to obtain a specific cost of 0 €/t<sub>CO2</sub>. Nevertheless, with these conditions the CAPEX is not amortized. To have a payback of 20 years, a specific carbon capture cost of -13 €/t<sub>CO2</sub> is required (i.e., actual benefits). Given a CO<sub>2</sub> tax of 162 €/t<sub>CO2</sub> and an electricity price of 35 €/MWh (wind power cost of production [52]), the CC cost would be -13 €/t<sub>CO2</sub>, the CAPEX would be amortized and the investment would be profitable.

#### 5. Conclusions

A novel concept that integrates Power to Gas technology through direct methanation in an oxy-blast furnace (OBF) was presented in this study. First, a PEM electrolyser produces green H<sub>2</sub> with renewable electricity, later, this green gas is mixed with some BFG in a methanation stage (direct BFG methanation), producing synthetic natural gas (SNG). The synthetic gas is ultimately injected into the OBF as a reducing agent, closing the carbon loop, reducing CO<sub>2</sub> emissions, and avoiding the use of additional fossil fuels. A study on the stoichiometry of the methanation reactor was performed, varying the H<sub>2</sub>:CO<sub>2</sub> ratio from 2.5 mol<sub>H2</sub>/mol<sub>CO2</sub>, going through 4 mol<sub>H2</sub>/mol<sub>CO2</sub>, and reaching an infinite ratio (full H<sub>2</sub>, no BFG).

For a low H<sub>2</sub>:CO<sub>2</sub> ratio of 2.5 poor results are found. In this configuration, less H<sub>2</sub> than BFG is injected in the methanation stage, and as a result a certain amount of CO<sub>2</sub> is directly injected into the OBF. This results in a severe increase in the coke rate in the OBF, which leads to a very low CO<sub>2</sub> emission reduction of 5%. As less H<sub>2</sub> is needed, the PEM size is also smaller, and an ASU is required to produce additional oxygen. Despite the lower PEM size, the specific CC cost increases, reaching 1518  $\ell/t_{\rm CO2}$ . The specific CC cost referred to HRC production is still relatively high, with 155  $\ell/t_{\rm HRC}$ .

For a stoichiometric H<sub>2</sub>:CO<sub>2</sub> ratio of 4, the SNG is composed mostly of CH<sub>4</sub> and is injected into the OBF. The coke rate decreases to 280 kg/ t<sub>HM</sub> leading to a CO<sub>2</sub> emission reduction of 33.8 %. A total of 443 kg/t<sub>HM</sub> of BFG are diverted to the methanation stage, the PEM size is 972 MW<sub>e</sub> and no ASU is needed. The specific CC costs of this configuration are 260  $\ell/t_{CO2}$  and 171  $\ell/t_{HRC}$ .

For full H<sub>2</sub> injection into the OBF, similar results are found with a stoichiometric H<sub>2</sub>:CO<sub>2</sub> ratio of 4. The coke rate increases slightly to 307 kg/t<sub>HM</sub>, resulting in a CO<sub>2</sub> emission reduction of 28.6 %. The PEM size is 930 MW and the H<sub>2</sub> produced is directly injected into the OBF, so no BFG is diverted to the methanation stage. The PEM also by-produces enough oxygen and the ASU is not required. The specific CC costs when full H<sub>2</sub> injection are 245  $\ell/t_{CO2}$  and 136  $\ell/t_{HRC}$ .

This PtG integration in the steelmaking industry recycles  $CO_2$  and CO via BFG methanation (except for full  $H_2$  injection), and is able to supply all downstream energy requirements (the power plant, sintering, etc.), as opposed to some TGR configurations that only recycle CO.

In general, the PtG-steelmaking integration with direct BFG methanation and oxy-blast furnace had the advantage of reducing  $CO_2$  emissions through carbon recycling. The use of low  $H_2:CO_2$  ratios is not recommended, as poor results are obtained.  $H_2:CO_2$  ratio of 4 and direct injection of  $H_2$  obtain similar results, with higher capture ratios in the former and better economic results in the latter.

# CRediT authorship contribution statement

Jorge Perpiñán: Conceptualization, Methodology, Writing – original draft, Visualization. Manuel Bailera: Conceptualization, Methodology, Writing – review & editing, Supervision, Project administration, Funding acquisition. Begoña Peña: Conceptualization, Methodology, Writing – original draft, Writing – review & editing, Visualization, Supervision, Project administration, Funding acquisition.

### **Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

# Data availability

Data will be made available on request.

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# Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.enconman.2023.117611.

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