

## Research paper

## Decarbonisation of lime kilns in ironmaking plants through Power to Gas

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## ABSTRACT

The iron and steel industry is one of the world's major carbon emitters. Various decarbonisation options are currently being explored: Asian producers mainly focus on carbon capture and carbon recycling in blast furnaces, while European producers favour the use of hydrogen in direct reduction processes. While the latter can eliminate CO<sub>2</sub> emissions from iron ore reduction, producers still face CO<sub>2</sub> emissions in other plant processes. Lime kilns, which are essential to iron production, cannot be fully decarbonized through hydrogen or direct electrification alone, as CO<sub>2</sub> is inherently released during the calcination of limestone. Consequently, this paper analyses different decarbonisation pathways for lime kilns in ironmaking plants. These pathways utilize Power to Gas to establish carbon recycling concepts to avoid CO<sub>2</sub> emissions. The integration of Power to Gas was modelled and simulated for the following lime kiln technologies: (1) Parallel flow regenerative shaft air-fuel lime kiln with amine scrubbing, (2) Parallel flow regenerative shaft oxy-fuel lime kiln, and (3) Parallel flow regenerative shaft oxy-fuel H<sub>2</sub> lime kiln. The Power to Gas technology is powered by solar power. The PV solar field was modelled as being installed on the rooftops of a steelworks located in Austria. The total PV capacity that can currently be installed is 15 MWp. When considering additional roofs that require moderate rehabilitation or cleaning, the total PV capacity increases to 57 MWp. The results show that CO<sub>2</sub> emissions can be reduced by 2% to 28% compared to the conventional air-blown lime kiln case. However, CO<sub>2</sub> avoidance costs exceed 1,000 €/t<sub>CO2</sub>, rendering the concept economically unfeasible.

1. Introduction: CO<sub>2</sub> emissions in iron and steel plants when shifting from coal to H<sub>2</sub>

A conventional ironmaking plant has numerous CO<sub>2</sub> sources (Fig. 1), which arise from the use of coal, natural gas and limestone as raw materials. Limestone is calcined to obtain lime, which produces CO<sub>2</sub> (Eq. (1)). Lime has various uses within the steel industry as: (1) a binder in sinter production, (2) a desulfurizing agent, (3) a slag forming component for removing impurities, and (4) a component to control melt chemistry and reduce damage to furnace linings. The heat for calcination is supplied by the combustion of fossil fuels, such as natural gas, which leads to additional CO<sub>2</sub> emissions (Eq. (2)). The CO<sub>2</sub> content in the flue gas of a lime kiln is around 20vol% [1] (Table 1).

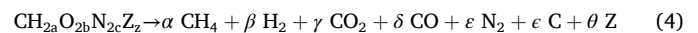


Coke is used in the sintering process, within coke ovens, and as an auxiliary fuel in blast furnaces. In sintering, coke is burned to

agglomerate iron ore fines with other fine materials (under 10 mm in size) to produce an open-grained, consistent material with a definite basicity, which is optimal for the blast furnace burden. In addition, sintering allows for the recycling of dust and ferrous materials by fusing them at high temperatures with iron ore fines, with little change to their chemical properties. The coke burned in the sinter strand produces CO<sub>2</sub> (Eq. (3)), which is emitted within the process flue gas. The CO<sub>2</sub> content of the sinter strand flue gas is around 8vol% [8].



In the coke oven, coal is heated to 1100 °C in an oxygen-free atmosphere (a carbonization process) to drive off volatile matter and produce coke, which is a solid residue consisting primarily of carbon (Eq. (4)). The CO<sub>2</sub> content of the process flue gas is low, at about 2vol% [9]. However, since coke oven gas is used as a fuel in various other parts of the ironmaking plant, its subsequent combustion results in significant CO<sub>2</sub> emissions.



Coke is the most critical raw material in the blast furnace. It acts as

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Nomenclature		$w$	Specific electricity consumption, kWh/Nm <sup>3</sup>
<b>Acronyms</b>		$\dot{W}$	Power, kW
BFG	Blast furnace gas	<b>Greek symbols</b>	
BOFG	Basic oxygen furnace gas	$\delta$	calculation parameter, h
CO <sub>2</sub> -eq	CO <sub>2</sub> equivalent emissions	$\epsilon$	conversion efficiency, Nm <sup>3</sup> <sub>SNG</sub> /Nm <sup>3</sup> <sub>H<sub>2</sub></sub>
COG	Coke oven gas	$\eta$	efficiency, -
CSP	Concentrated solar power	$\nu$	stoichiometry of methanation, mol <sub>H<sub>2</sub></sub> /(mol <sub>CO<sub>2</sub></sub> +mol <sub>CO</sub> )
I&S	Iron and steel	$\tau$	storage capacity in terms of nominal operating hours, h
LDC	Load duration curve	<b>Subscripts and superscripts</b>	
LHV	Lower heating value	*	Nominal load / full capacity
PCI	Pulverized coal injection	#	Partial load
PFR	Parallel flow regenerative (shaft kiln)	-	Dropped / discarded
PtG	Power to gas	1	Interval of 1-hour duration
PV	Photovoltaic	$b$	Li-ion battery
PVGIS	Photovoltaic geographical information system	$e$	Electrolyser
SNG	Synthetic natural gas	$i$	Lime kiln configuration $i$
THM	Ton of hot metal	$j$	Hour $j$ of the year
<b>Symbols</b>		$m$	methanation
$t$	Operating hours, h	$p$	Peak power
$V$	Volume produced/consumed/available, Nm <sup>3</sup>	$PV$	Photovoltaic panels
$W$	Electricity, kWh	$s$	H <sub>2</sub> storage / H <sub>2</sub> tank

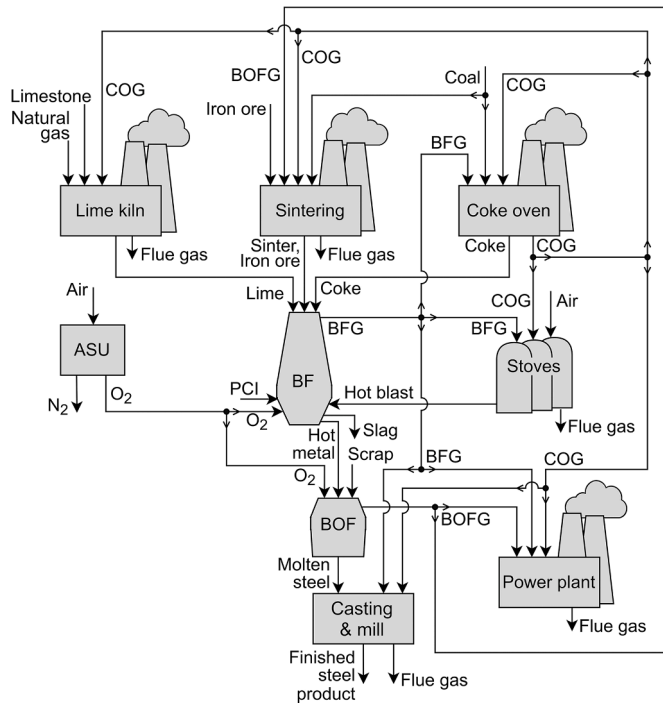


Fig. 1. Example of a process flow diagram of an integrated I&S plant (not all plants have the same configuration).

reducing agent to reduce iron ore directly (Eq. (5)) and indirectly (via the CO obtained from coke combustion, Eqs. (6) to (8)). Coke also provides mechanical stability and permeability inside the furnace, ensuring the ascent of the reducing gases, as well as optimal resource utilization and high productivity. Pulverized coal injection can be used as an auxiliary fuel in the lower part of the furnace to decrease coke

Table 1  
Typical composition (vol%) of flue gases in an I&S plant [2-7].

Flue gas from	CO <sub>2</sub>	CO	H <sub>2</sub> O	H <sub>2</sub>	CH <sub>4</sub>	C <sub>2</sub> H <sub>4</sub>	N <sub>2</sub>	O <sub>2</sub>
Lime kiln	15 – 25	-	5 – 15	-	-	-	50 – 70	1 – 9
	1 – 8	0 – 1	2 – 10	-	-	-	66 – 77	15 – 20
Sintering	1 – 3	4 – 8	0 – 4	39 – 65	20 – 42	0 – 2	2 – 8	0 – 1
	17 – 25	20 – 28	2 – 3	1 – 5	-	-	46 – 55	-
Coke oven	20 – 30	-	5 – 15	-	-	-	52 – 69	0 – 1
	14 – 16	57 – 73	0 – 12	0 – 3	-	-	8 – 14	-
Blast furnace	10 – 30	0 – 2	5 – 20	-	-	-	50 – 90	0 – 5
	15 – 20	-	5 – 15	-	-	-	50 – 80	0 – 7
Stoves	15 – 20	-	5 – 15	-	-	-	50 – 80	0 – 7
	15 – 20	-	5 – 15	-	-	-	50 – 80	0 – 7
BOF	15 – 20	-	5 – 15	-	-	-	50 – 80	0 – 7
	15 – 20	-	5 – 15	-	-	-	50 – 80	0 – 7
Casting & mill	15 – 20	-	5 – 15	-	-	-	50 – 80	0 – 7
	15 – 20	-	5 – 15	-	-	-	50 – 80	0 – 7
Power plant	15 – 20	-	5 – 15	-	-	-	50 – 80	0 – 7
	15 – 20	-	5 – 15	-	-	-	50 – 80	0 – 7

consumption. The top gas produced in the blast furnace mainly consists of CO<sub>2</sub> from the reduction reactions, unreacted CO, and N<sub>2</sub> from the blast air. The CO<sub>2</sub> content is around 23vol%. Similar to coke oven gas, this blast furnace gas can be used as a fuel in other plant processes, which results in further CO<sub>2</sub> emissions. One of the main users of blast furnace gas is the hot blast stoves, which use this fuel gas to preheat the air entering the blast furnace.



Another process that emits CO<sub>2</sub> is the basic oxygen furnace. Here, the

carbon content of the hot metal is diminished by oxidation to obtain steel (Eq. (9)). Part of the CO produced in the process is also oxidised to CO<sub>2</sub> because of the presence of oxygen (Eq. (10)). The CO<sub>2</sub> content of the BOF gas is around 16vol%. The carbon that is dissolved in the hot metal came from the coke used in the blast furnace, which in turn came from the coal used in the coke oven.



During the casting of the steel, mainly coke-oven gas and some blast-furnace gas are used for reheating purposes. The exhaust gas has a low CO<sub>2</sub> content (7vol%) due to the high H<sub>2</sub> content of the coke oven gas [10]. The remaining fuel gases are used to generate electricity in a power plant. The most common technology is combined cycle, whose exhaust gas typically contains 17% CO<sub>2</sub> for this type of fuel gases.

Attending to all the CO<sub>2</sub>-emitting processes of iron and steel plants, one realises that coal is behind most of them, driven by the necessity of coke in the blast furnace. In the near future, this scenario may drastically change with the adoption of new technologies based on the direct reduction of iron ore by H<sub>2</sub> (Eqs. (11) to (13)). Direct reduction refers to solid-state processes that reduce iron ore to metallic iron at temperatures below the melting point (below 1200 °C). The iron obtained this way is called direct reduced iron (DRI) or sponge iron [11]. If the direct-reduced iron is compacted at densities above 5,000 kg/m<sup>3</sup>, it is considered a premium form of DRI and called hot briquetted iron (HBI).



The utilisation of shaft furnaces for DRI, instead of blast furnaces, will avoid the emissions of the latter, but also the emissions of coke making, which will no longer be needed. At the same time, those processes using blast furnace gas and coke oven gas will shift to H<sub>2</sub> to fulfil their heat requirements. In addition, the power plant might disappear if there are no fuel gases to take advantage of, thus being replaced by direct renewable electricity (wind or solar power). In the case of sintering, the palletisation of iron ore will still be required for DRI, for which some solid fuel will have to be consumed. Recently, Vale (mining company) produced iron ore pellets using plant-based biochar for the first time, instead of anthracite coal [12]. Moreover, other biomass fuels have been studied from a theoretical point of view by Sefidari et al [13], who concluded that pyrolysis oils could be the most viable alternative to coal in the pelletizing processes. Therefore, it is very likely that pelletizing will switch to CO<sub>2</sub>-neutral fuels in the future as well. On its part, the basic oxygen furnace will be replaced by an electric arc furnace (EAF) to achieve the required steel grade through carburization [14]. In the EAF, DRI and/or HBI are charged together with alloying elements, slag formers (such as lime) and carbon sources to control the carbon content of the melt. As for pelletisation, some authors already studied the possibility of using biomass or biochar as carbon sources for the EAF process [15]. Therefore, any CO and CO<sub>2</sub> in the exhaust gas of the EAF will come from CO<sub>2</sub>-neutral fuels. Lastly, the lime kiln, which is necessary for slag control, may run on renewable fuels to provide the heat for calcination. However, calcination of limestone will always produce CO<sub>2</sub> that does not come from CO<sub>2</sub>-neutral fuels. Still, from the limestone itself (Eq. (1)). Thus, when in the near future steel producers shift from coal to H<sub>2</sub>, they will be able to avoid all CO<sub>2</sub> emissions except for those coming from lime production.

The objective of the present paper is to propose and analyse four different pathways for decarbonising lime kilns in ironmaking plants. These concepts are based on methanation, which produces synthetic methane by combining CO<sub>2</sub> with renewable H<sub>2</sub> (Eq. (14)).



The novelty of the present papers lies in the fact that none of the proposed concepts has been analysed before in the context of a future H<sub>2</sub>-based steelmaking industry, where the only remaining emissions are those from lime kilns. Moreover, some of the proposed concepts are entirely new and can be extrapolated to other industries (e.g., cement industry). All the proposed solutions are based on commercially available or under-research technologies in industrial pilot plants. In addition, the required renewable electricity is assumed to come from photovoltaic panels installed in the different buildings of an ironmaking plant located in Austria, by quantifying the available roof with Google Earth. The paper is completed with the techno-economic analysis of the proposed solutions. The study comprises: (1) the description of the decarbonisation technologies proposed, (2) the methodology on both the PV solar field and the methanation plant sizing and management, (3) the methodology for simulating the different decarbonisation technologies, (4) the methodology on techno-economic analysis, (5) the assessment on the renewable electricity and H<sub>2</sub> production under two different scenarios, (6) the assessment on CO<sub>2</sub> emissions reduction, and (7) the analysis of cash flow, pay-back and internal rate of return of the concepts.

## 2. Decarbonisation pathways based on Power to Gas for lime kilns

Calcination is a chemical reaction that dissociates limestone (CaCO<sub>3</sub>) into lime (CaO) and CO<sub>2</sub> by consuming thermal energy at high temperature (>850 °C). This CO<sub>2</sub> must be consumed, recycled or stored to avoid its release into the atmosphere. In our study, we opted for methanation (Eq. (14)) since hydrogen will be produced in large

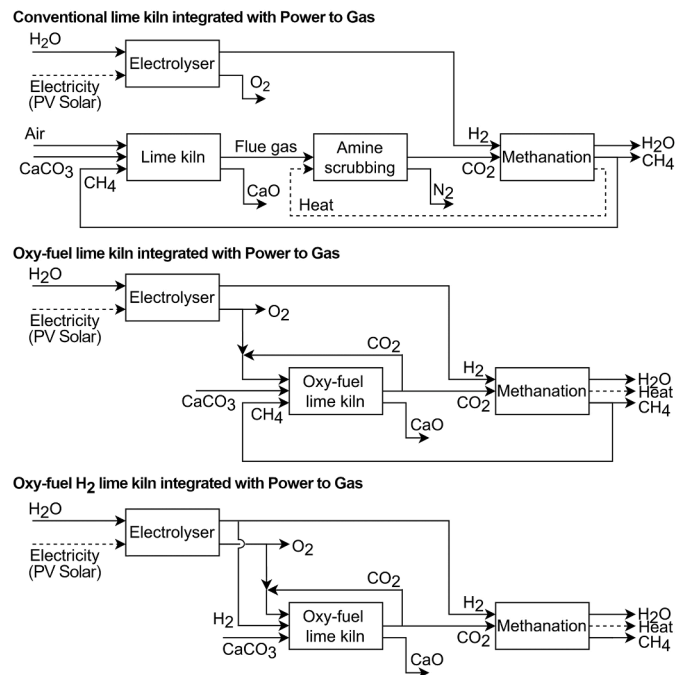


Fig. 2. Decarbonisation options based on Power to Gas for lime kilns.

quantities in a future H<sub>2</sub>-based DRI steelmaker. Additionally, the thermal energy required for calcination should be provided by CO<sub>2</sub>-neutral synthetic fuels, hydrogen or even concentrated solar power, to avoid further emissions. Considering these premises, three decarbonisation options were analysed. These are presented in Fig. 2, considering that all fossil natural gas is replaced and that all CO<sub>2</sub> is sent to methanation. In practice, reliance on some fossil fuels and/or the inability to fully utilise CO<sub>2</sub> may persist if renewable electricity proves insufficient. Additionally, hydrogen availability may remain limited in the medium term.

- **Conventional lime kiln integrated with amine scrubbing and Power to Gas:** The first decarbonisation option combines air-blown lime kilns (conventional technology) with amine scrubbing to capture the CO<sub>2</sub> emissions. Then, the captured CO<sub>2</sub> is sent to the methanation stage, where it is combined with renewable hydrogen and produce synthetic methane. Part of this synthetic methane is used in the lime kiln to replace the fossil natural gas. In addition, the amine scrubbing takes advantage of the exothermic heat from methanation to provide heat to the reboiler of the CO<sub>2</sub> desorption tower.
- **Oxy-fuel lime kiln integrated with Power to Gas:** The second decarbonisation option is an oxy-fuel lime kiln. This uses a mixture of pure O<sub>2</sub> and recirculated flue gas as an oxidising agent in combustion, instead of air. The recirculated CO<sub>2</sub> replaces the N<sub>2</sub> in the air to maintain operating conditions like those of conventional technologies. Thanks to the oxy-fuel conditions, CO<sub>2</sub> concentrations above 90vol% can be achieved in the exhaust gas, thus avoiding amine scrubbing [16]. Moreover, oxy-fuel lime kilns can take advantage of the O<sub>2</sub> produced during the water electrolysis stage. As in the previous decarbonisation option, CO<sub>2</sub> is used to produce synthetic methane, which replaces the fossil natural gas in the lime kiln.
- **Oxy-fuel H<sub>2</sub> lime kiln integrated with Power to Gas:** The third decarbonisation option is also an oxy-fuel lime kiln, like in the previous case, but now hydrogen is used as fuel. The CO<sub>2</sub> from calcination is used to perform methanation. Since this lime kiln does not consume natural gas, the synthetic methane is injected into the grid for sale.

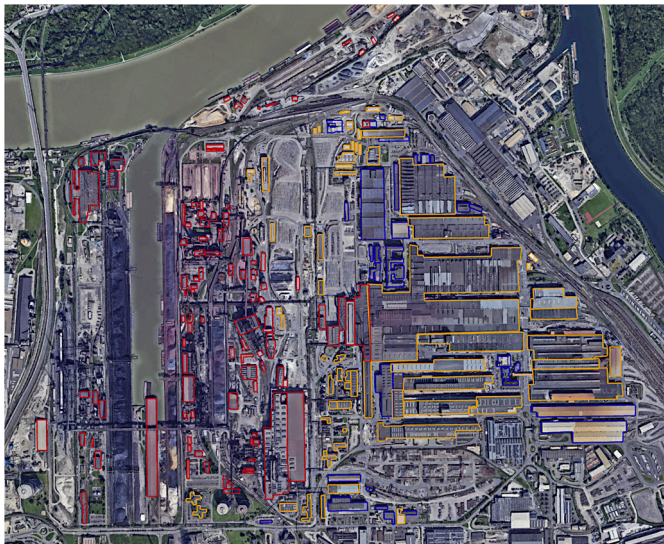


Fig. 3. Satellite image for a steelworks located in Austria (48°16'38.2"N, 14°19'58.7"E) [20]. Blue: Type 1 roof, Yellow: Type 2 roof, Red: Type 3 roof.

### 3. Methodology

#### 3.1. Sizing and management of the PV solar field

The PV solar field was assumed to be installed on the roofs of the various buildings at a steelworks in Austria (Fig. 3). The available roof area was delimited in Google Earth and quantified in QGIS. The roofs were classified into three categories:

- Type 1: These roofs are assumed to be ready for PV panel installation, and their operation would not be notably affected by surrounding conditions (shadows, dust, etc.).
- Type 2: These roofs need conditioning to install PV panels and/or moderate measures to mitigate ambient dust from nearby processes. Hence, additional investment would be required.
- Type 3: These roofs require remarkable modifications and/or extreme measures against the ambient dust from nearby processes, hence jeopardising the economic viability of the PV panels.

The electricity production from the PV panels was calculated using the PVGIS tool from the European Commission (version 5.2) and the PVGIS-SARAH2 database [17]. The mounting type was assumed fixed, and the PVGIS tool optimised the panels' slope and azimuth. The PV technology was crystalline silicon, and the system loss was fixed at 20%, which is higher than the default value of 14% because of the expected ambient dust in the steelworks (it must be noted that this loss does not include losses related to the angle of incidence and temperature, which are already estimated and taken into account by the PVGIS tool according to the satellite data). The data on electricity production were retrieved hourly for 1 kWp of power capacity, at latitude 48.271 and longitude 14.336, for the year 2019 (Table 2). This electricity production was then scaled up to the total peak power that can be installed in the available solar area by assuming 15% efficiency [18,19] of the PV panels (Eq. (15)). The available solar area is assumed 70% for Type 1 roof, 50% for Type 2 roof, and 0% for Type 3 roof.

$$\text{Peak power [kWp]} = \text{Solar area [m}^2\text{]} \times \text{PV panel efficiency [-]} \quad (15)$$

Additionally, a Li-ion battery was included to mitigate variations in the electricity production from the PV panels, which might occur due to cloud coverage. The nominal power of the battery ( $\dot{W}_b$ ) was assumed to be 5% of the total peak power of the PV solar field ( $0.05\dot{W}_{PV,p}^*$ ) [21], the efficiency of the battery ( $\eta_b$ ) was set at 85% [22,23], and its energy

Table 2

Location and technical data of the PV solar field.

Parameter	Data
Location	Linz, Austria
Latitude (dec)	48.271
Longitude (dec)	14.336
Type 1 roof	
N° buildings	30
Roof area (m <sup>2</sup> )	141,048
Solar area (m <sup>2</sup> )	98,734
Type 2 roof	
N° buildings	64
Roof area (m <sup>2</sup> )	563,053
Solar area (m <sup>2</sup> )	281,527
Type 3 roof	
N° buildings	101
Roof area (m <sup>2</sup> )	221,130
Solar area (m <sup>2</sup> )	0
PV technology	Crystalline silicon
PV system loss (%)	20
PV panel efficiency (%)	15
PV panel slope (°)	38
PV panel azimuth (°)	1

capacity equal to 4 h of nominal operation ( $\tau_b$ ) [21,22]. The electricity available in the Li-ion battery at hour  $j$  ( $W_{b,j}$ ) depends on the PV electricity production ( $W_{PV,j-1}$ ) and the charge of the battery ( $W_{b,j-1}$ ) at the preceding hour  $j-1$ , and on the electrolyser consumption at nominal load for 1 hour ( $W_{e,1}^*$ ) (Eq. (16)).

$$W_{b,j} = \begin{cases} W_b^* & \text{if } W_{PV,j-1} \geq W_{e,1}^* \text{ and } \left( (W_{PV,j-1} - W_{e,1}^*)\eta_b + W_{b,j-1} \right) \geq W_b^* \\ (W_{PV,j-1} - W_{e,1}^*)\eta_b + W_{b,j-1} & \text{if } W_{PV,j-1} \geq W_{e,1}^* \text{ and } \left( (W_{PV,j-1} - W_{e,1}^*)\eta_b + W_{b,j-1} \right) < W_b^* \\ (W_{PV,j-1} + W_{b,j-1}) - W_{e,1}^* & \text{if } W_{PV,j-1} < W_{e,1}^* \text{ and } (W_{PV,j-1} + W_{b,j-1}) \geq W_{e,1}^* \\ 0 & \text{if } W_{PV,j-1} < W_{e,1}^* \text{ and } (W_{PV,j-1} + W_{b,j-1}) < W_{e,1}^* \end{cases} \quad (16)$$

It should be noted that the available electricity in the battery is accounted for after losses. At full capacity ( $W_b^*$ ), it is equal to its nominal power capacity ( $\dot{W}_b^*$ ) multiplied by the equivalent hours of energy storage ( $\tau_b$ ), accounting for losses (i.e.,  $W_b^* = \tau_b \eta_b \dot{W}_b^*$ ).

### 3.2. Sizing and management of the Power to Gas plant

Once both the electricity produced by the PV panels (data from PVGIS scaled by Eq. (15)) and the electricity provided by the Li-ion battery (Eq. (16)) were known, the electricity consumed by the electrolyser was computed through Eq. (17).

$$W_{e,j} = \begin{cases} W_{e,1}^* & \text{if } W_{PV,j} \geq W_{e,1}^* \\ W_{e,1}^* & \text{if } W_{PV,j} < W_{e,1}^* \text{ and } (W_{PV,j} + W_{b,j}) \geq W_{e,1}^* \\ W_{PV,j} + W_{b,j} & \text{if } (W_{PV,j} + W_{b,j}) < W_{e,1}^* \end{cases} \quad (17)$$

In order to select  $W_{e,1}^*$  (which is the nominal electricity consumption of the electrolyser in 1 hour,  $W_{e,1}^* = 1 \cdot W_e^*$ ), the electrolyser was sized using the load duration curve (LDC) (Fig. 4). This curve relates the nominal operating load of the electrolyser ( $\dot{W}_e^*$ ) with the maximum operating hours that it might work at that load ( $t_{\dot{W}_e^*}$ , Eq. (18)).

$$t_{\dot{W}_e^*} = \sum_j \delta_j \quad \text{where} \quad \delta_j = \begin{cases} 0 & \text{if } W_{e,j} < W_{e,1}^* \\ 1 & \text{if } W_{e,j} = W_{e,1}^* \end{cases} \quad (18)$$

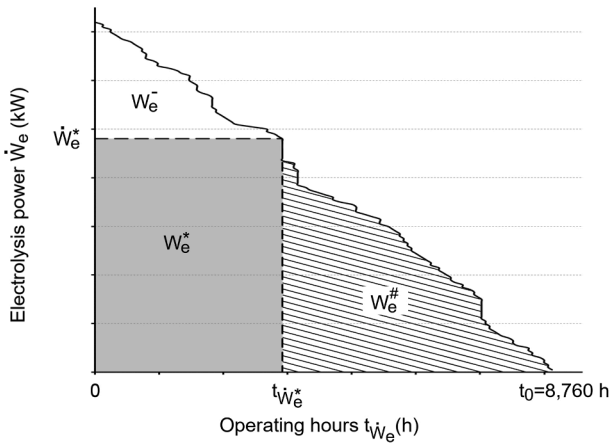


Fig. 4. Load duration curve methodology (blank area: energy discarded; grey area: energy processed at nominal load; pattern area: energy processed at partial load).

The product of the two values ( $\dot{W}_e^* \cdot t_{\dot{W}_e^*}$ ) is a measure of the electrical energy that could be processed by the electrolyser at nominal load ( $W_e^*$ ). The main criterion (criterion 1) for selecting the nominal power capacity of the electrolyser was to maximise the energy processed at nominal load (i.e., maximisation of  $W_e^*$ , the grey area under the LDC, in Fig. 4) [24]. In some cases, large variations in the resulting nominal load (e.g.,

$\dot{W}_e^* \pm 15\%$ ) only produced small changes in the energy processed at nominal load ( $W_e^* \pm 0.1\%$ ), but the total energy processed remarkably changed ( $(W_e^* + W_e^\#) \pm 6\%$ ). Hence, in these cases, a second criterion (criterion 2) was added to avoid ambiguity: the nominal load derived from the maximisation process was increased by progressively decreasing  $W_e^*$  at  $-0.25\%$  intervals until the relative increment in the total processed energy dropped below 1%. Graphically, this means that the grey area ( $W_e^*$ ) was diminished at  $-0.25\%$  intervals by increasing  $W_e^\#$  in Fig. 4, until the total area composed by  $W_e^* + W_e^\#$  stopped increasing beyond 1% in a single step. This way, we had an electrolyser with a very similar operational optimisation (nearly the same amount of energy processed at nominal load), but with a notable greater total energy processed (i.e., the blank area of discarded energy  $W_e^-$  notably decreased, the pattern area of energy processed at partial load  $W_e^\#$  increased in the same amount, and the grey area of energy processed at nominal load  $W_e^*$  barely changed).

The amount of  $H_2$  produced by the electrolyser was calculated using Eq. (19), which is directly dependent on the electricity supplied. The specific consumption,  $w_e$ , was set at  $4.5 \text{ kWh/Nm}^3$ , corresponding to a commercial containerised PEM electrolyser in the M scale [25].

$$V_{H_2,e,j} = W_{e,j} / w_e \quad (19)$$

This  $H_2$  was either sent to storage or to methanation, depending on the  $H_2$  hourly production of the electrolyser ( $V_{H_2,e,j}$ ) and on the nominal  $H_2$  consumption of the methanation plant ( $V_{H_2,m,1}^*$ ). The  $H_2$  available in the tank at each hour was calculated using Eq. (20), and the  $H_2$  consumption of the methanation plant was calculated using Eq. (21).

$$V_{H_2,s,j} = \begin{cases} V_{H_2,e,j-1} + V_{H_2,s,j-1} - V_{H_2,m,1}^* & \text{if } (V_{H_2,e,j-1} + V_{H_2,s,j-1}) > V_{H_2,m,1}^* \\ 0 & \text{if } (V_{H_2,e,j-1} + V_{H_2,s,j-1}) \leq V_{H_2,m,1}^* \end{cases} \quad (20)$$

$$V_{H_2,m,j} = \begin{cases} V_{H_2,m,1}^* & \text{if } (V_{H_2,e,j} + V_{H_2,s,j}) \geq V_{H_2,m,1}^* \\ V_{H_2,e,j} + V_{H_2,s,j} & \text{if } (V_{H_2,e,j} + V_{H_2,s,j}) < V_{H_2,m,1}^* \end{cases} \quad (21)$$

The Eq. (20) assumes no limit on the  $H_2$  storage, so the required size for the  $H_2$  tank is  $\max_j(V_{H_2,s,j})$ . This value is entirely dependent on methanation plant size: the smaller the methanation plant, the larger the  $H_2$  tank (unsuitable for technical and economic viability) and the higher the methanation plant's capacity factor (suitable for steady operation). Hence, we minimised the methanation plant size in order to increase its equivalent operating hours, subject to the constraint:  $\max_j(V_{H_2,s,j}) \leq 66,740$  (constrained minimisation solver). This limit is intended for the storage of  $6,000 \text{ kg of } H_2$  (i.e.,  $66,740 \text{ Nm}^3$ ), which corresponds to the  $H_2$  storage installed in Puertollano (Spain) by Iberdrola at a 100 MW PV

solar plant aimed at producing of green H<sub>2</sub>.

The H<sub>2</sub> is stored at 60 bar in 11 tanks, each 133 m<sup>3</sup> each (23.5 m in height, 2.8 m in diameter and 4.5 cm in thickness) [21]. The SNG production (Eq. (22)) and the CO<sub>2</sub>-eq consumption (Eq. (23)) will depend on the lime kiln configuration selected, due to differences in flue gas composition.

$$V_{SNG,m,j} = \begin{cases} V_{H_2,m,1}^* \epsilon_{m,i} & \text{if } (V_{H_2,e,j} + V_{H_2,s,j}) > V_{H_2,m,1}^* \\ (V_{H_2,e,j} + V_{H_2,s,j}) \epsilon_{m,i} & \text{if } (V_{H_2,e,j} + V_{H_2,s,j}) \leq V_{H_2,m,1}^* \end{cases} \quad (22)$$

$$V_{CO_2\text{-eq},m,j} = \begin{cases} V_{H_2,m,1}^* / \nu_{m,i} & \text{if } (V_{H_2,e,j} + V_{H_2,s,j}) > V_{H_2,m,1}^* \\ (V_{H_2,e,j} + V_{H_2,s,j}) / \nu_{m,i} & \text{if } (V_{H_2,e,j} + V_{H_2,s,j}) \leq V_{H_2,m,1}^* \end{cases} \quad (23)$$

The parameters  $\epsilon_{m,i}$  (gas conversion efficiency) and  $\nu_{m,i}$  (stoichiometry of methanation) were calculated in Aspen Plus for each configuration.

### 3.3. Modelling of the lime kiln and Power to Gas plants

#### 3.3.1. Air-blown lime kiln with amine scrubbing

The first decarbonisation option combines an air-blown lime kiln and amine scrubbing. The calcination technology used in steelworks is normally a PFR kiln (parallel-flow regenerative shaft kiln). It consists of two vertical shafts and a connecting crossover channel (Fig. 5). Both shafts work together: one calcines the product and the other preheats the stone. In the burning shaft, the lime is calcined in parallel-flow system. The hot combustion gases are then transferred through the crossover channel to the non-burning shaft, where they preheat the limestone in a counterflow arrangement. The flow direction of the gases is reversed at regular intervals, alternating the function of the shafts [26].

The PFR kiln was modelled in Aspen Plus, assuming a typical lime production of 200 t/d. The burning shaft comprises a RGibbs reactor and a heat exchanger. The reactor limits the maximum CaCO<sub>3</sub> conversion to 98.5%, and considers Fe<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> as non-reacting components. The heat exchanger cools the lime with the air entering at the bottom, which is heated to 950 °C. The non-burning shaft accounts the heat transfer between the gases (exiting at 100 °C) and the limestone, during the preheating of the solids. The simulation used the PENG-ROB property method, based on the Standard Peng-Robinson cubic equation of state [27].

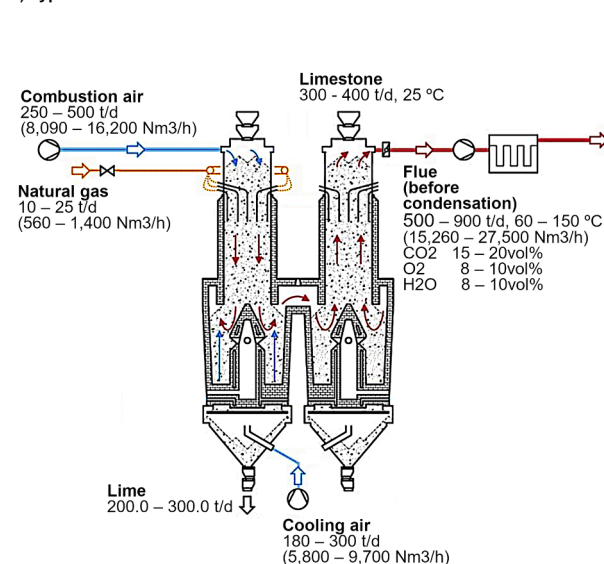
The amine scrubbing model was elaborated from an Example File provided by Aspen Plus for MDEA [28]. Before diverting the flue gas to the capture plant, water was condensed at 35 °C. The CO<sub>2</sub> capture ratio was fixed at 90%. The clean gas exits the absorber at 1 bar and 42 °C, while the CO<sub>2</sub>-rich gas exits the amine plant at 1 bar and 35 °C (after condensation) with a CO<sub>2</sub> content of 95.9vol%. The heat demand for the stripper reboiler is 3.6 MJ/kgCO<sub>2</sub>, which can be supplied with the exothermic heat from methanation. A detailed description of the amine plant is available in [29].

#### 3.3.2. Oxy-fuel lime kiln

The second decarbonisation option is an oxyfuel PFR kiln (Fig. 6). In this case, recirculation gas enriched with oxygen is used as an oxidising agent instead of air. In addition, the cooling air at the bottom is extracted from the kiln before it can mix with the combustion gas, thereby preventing N<sub>2</sub> from entering the flue gas. Recirculation gas is also used to recover the heat from the cooling air and then injected into the connecting channel (to replace the discharged cooling air), thus maintaining similar mass flow conditions. This technology, developed by Maerz, is known as the EcoKiln system [30]. This type of oxyfuel PFR kilns allows selecting both conventional air-blown operation and oxy-fuel operation, providing greater flexibility in terms of economic efficiency. In the oxyfuel mode, CO<sub>2</sub> concentrations above 90vol% in the exhaust gas, and lime productions of up to 800 t/d can be achieved [16].

The Aspen Plus model of the oxyfuel PFR considered the burning shaft (RGibbs reactor and heat exchanger for cooling) and the non-burning shaft (heat exchanger for preheating solids), as in the previous case, and included the necessary additional equipment for gas recirculation. To moderate the combustion temperature, the O<sub>2</sub> was mixed with flue gas. The amount of gas recirculated was fixed to obtain the same lime temperature at the outlet of the RGibbs reactor as in the air-blown PFR kiln. Additionally, the cooling air was discharged before entering the calcination stage (to avoid N<sub>2</sub> presence in the exhaust gas) and replaced with an equivalent recirculated mass flow through the connecting channel. The cooling air, which reached 950 °C after cooling the lime, is used to preheat the second recirculation to 750 °C. A further heating of the recirculation to 920 °C was necessary to prevent re-carbonisation of the already calcined product (this is usually performed using a small oxyfuel burner or an electric flow heater) [16]. The simulation used the PENG-ROB property method, based on the Standard Peng-Robinson cubic equation of state [31].

**AIR-BLOWN LIME KILN**  
a) Typical data



b) Aspen Plus simulation

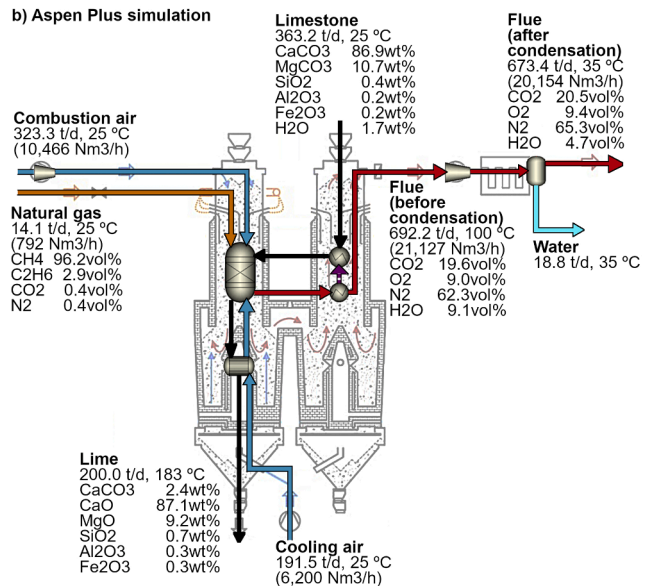


Fig. 5. Conceptual diagram (left) and Aspen Plus simulation (right) of an air-blown parallel flow regenerative lime kiln.

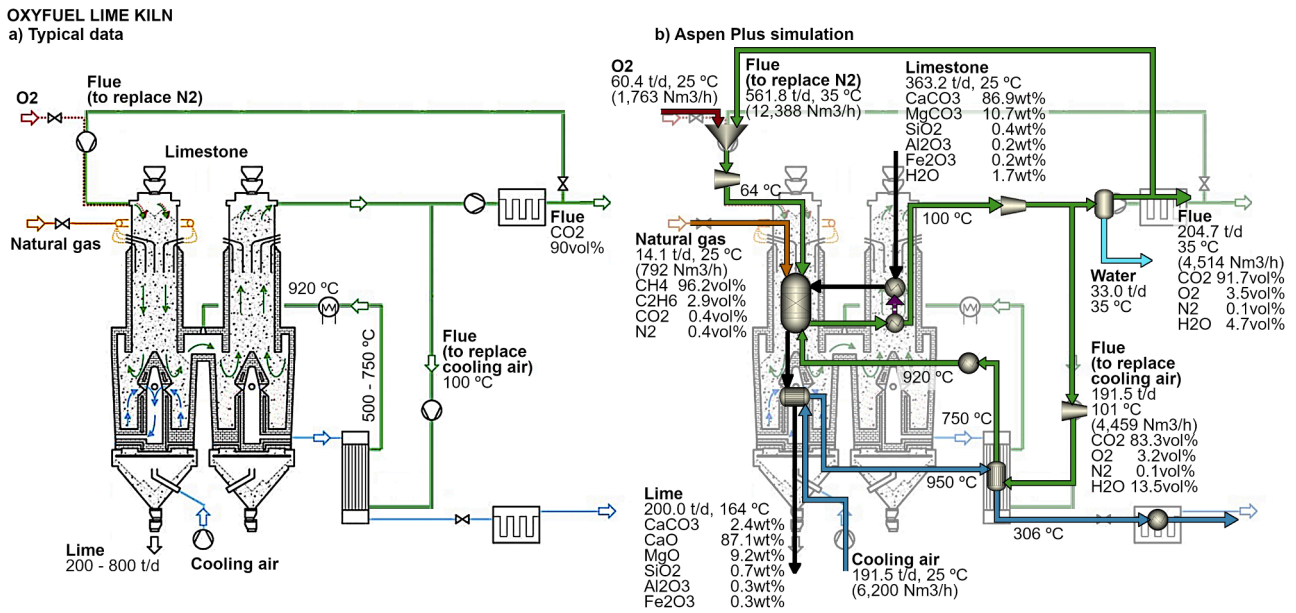


Fig. 6. Conceptual diagram (left) and Aspen Plus simulation (right) of an oxyfuel parallel flow regenerative lime kiln.

3.3.3. Oxy-fuel H<sub>2</sub> lime kiln

The third decarbonisation option is an oxyfuel PFR kiln, as in the previous case, but fuelled with H<sub>2</sub> instead of natural gas. The assumptions on temperatures and recirculation, as well as the property methods and models selected in Aspen Plus, are the same as in the previous case. The process flow diagram is presented in Fig. 7.

3.3.4. Methanation plant

The methanation plant is based on the technology developed by Hitachi Zosen Corporation. Their technology comprised two shell-and-tube typed exchange reactors, operating at 5 bar and 250 °C, with an intermediate condensation stage. The final CH<sub>4</sub> content of the synthetic natural gas was 98.5vol%, and the H<sub>2</sub> content was 1.3vol%, on a dry basis [32]. The corresponding Aspen Plus model consisted of two 2-stage compressors for the inlet CO<sub>2</sub> and H<sub>2</sub> (compression ratios of 2.5:1 and 2:1, with intermediate cooling at 60 °C), two RGibbs equilibrium

reactors for the methanation stages (at 250 °C and 5 bar), two Flash reactors for water condensation after each methanator (at 50 °C and 35 °C), and two preheating exchangers before each methanator (at 250 °C). Additionally, one RStoic reactor was placed upstream of the methanation plant, to catalytically combust any O<sub>2</sub> in the flue gas using H<sub>2</sub> (O<sub>2</sub> is combined with H<sub>2</sub> to produce H<sub>2</sub>O via catalysis rather than combustion) [33]. This additional stage is needed because O<sub>2</sub> deactivates the methanation catalyst. This is especially relevant for these decarbonisation concepts, since the flue gas contains around 3 - 9vol% O<sub>2</sub>. Therefore, the H<sub>2</sub> required for these concepts was slightly higher than expected from the stoichiometry of Eq. (14), to account for the O<sub>2</sub>.

3.4. Methodology for the economic analysis

The economic analysis considered capital expenditure (CAPEX), operational expenditure (OPEX) and income for the new and retrofitted

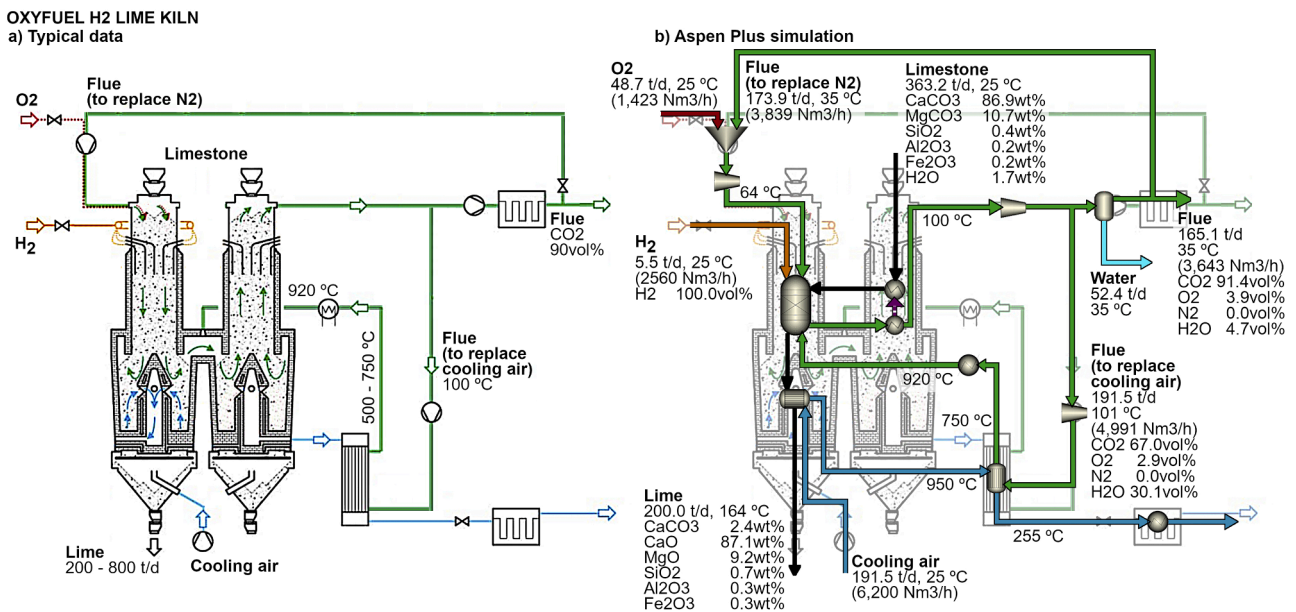


Fig. 7. Conceptual diagram (left) and Aspen Plus simulation (right) of an oxyfuel parallel flow regenerative H<sub>2</sub> lime kiln.

**Table 3**  
Data for the economic analysis.

	Cost equation (M€) or (M€/year)	Parameters $\alpha, \beta, \gamma$	Ref.
<b>CAPEX</b>			
Amine Plant	$43.49 \cdot (\alpha/408)^{0.65}$	CO <sub>2</sub> captured [t/h]	[36, 37]
Electrolysis	$1750 \cdot 10^{-6} \cdot \alpha$	Power [kW]	[38]
H <sub>2</sub> storage			
H <sub>2</sub> storage tank	$563 \cdot 10^{-6} \cdot \alpha$	H <sub>2</sub> mass [kg <sub>H2</sub> ]	[39]
H <sub>2</sub> compressor	$0.267 \cdot (\alpha/445)^{0.67}$	Power [kW]	[40]
<b>Methanation</b>			
CO <sub>2</sub> compressor	$0.267 \cdot (\alpha/445)^{0.67}$	Power [kW]	[40]
Reactors	$300 \cdot 10^{-6} \cdot \alpha$	SNG power [kW <sub>SNG</sub> ]	[41]
Catalyst	$0.1875 \cdot \alpha$	Volume of catalyst [m <sup>3</sup> ]	[40]
<b>Lime kiln</b>			
New lime kiln	$10^{-6} \cdot \alpha \cdot \beta$	New lime kiln price [€]	[42]
<b>Solar</b>			
PV plant	$750 \cdot 10^{-6} \cdot \alpha$	Power [kW]	-
Batteries	$500 \cdot 10^{-6} \cdot \alpha$	Battery size [kWh]	[43]
Thermal-solar field	$2344 \cdot 10^{-6} \cdot \alpha$	Solar field [kW]	[44]
<b>Other direct costs</b>			
Installation	$39\% \cdot \alpha$	Total equipment costs [€]	[45]
Instrumentation & control	$26\% \cdot \alpha$	Total equipment costs [€]	[45]
Piping	$31\% \cdot \alpha$	Total equipment costs [€]	[45]
Electrical	$10\% \cdot \alpha$	Total equipment costs [€]	[45]
Building	$29\% \cdot \alpha$	Total equipment costs [€]	[45]
<b>Indirect costs</b>			
Engineering	$32\% \cdot \alpha$	Total equipment costs [€]	[45]
Legal expenses	$4\% \cdot \alpha$	Total equipment costs [€]	[45]
Construction expenses	$34\% \cdot \alpha$	Total equipment costs [€]	[45]
Contingency	$37\% \cdot \alpha$	Total equipment costs [€]	[45]
<b>OPEX</b>			
Amine renovation	$3204 \cdot 10^{-6} \cdot \alpha$	MDEA renovation [tCO <sub>2</sub> /year]	[46]
Catalyst renovation	$15\% \cdot \alpha$	Initial catalyst cost [M€]	[35]
Electricity	$10^{-6} \cdot \alpha \cdot \beta$	Electricity cost [€/MWh], Electricity purchased [MWh/year]	-
Water	$1.47 \cdot 10^{-6} \cdot \alpha$	Water consumption [m <sup>3</sup> /year]	[47]
O&M	$3\% \cdot \alpha$	Total CAPEX	[36]
<b>INCOMES</b>			
Sold synthetic natural gas	$40 \cdot 10^{-6} \cdot \alpha$	SNG sold [MWh/year]	[48]
Saved fossil natural gas	$40 \cdot 10^{-6} \cdot \alpha$	NG saved [MWh/year]	[48]
Oxygen	$80 \cdot 10^{-6} \cdot \alpha$	O <sub>2</sub> generated [tO <sub>2</sub> /year]	[49]
CO <sub>2</sub> taxes	$84.28 \cdot 10^{-6} \cdot \alpha$	CO <sub>2</sub> consumed [tCO <sub>2</sub> /year]	[50]

components (Table 3). The CAPEX comprised the electrolyser, the methanation plant, the amine scrubbing/new lime kiln (depending on the decarbonisation pathway), the H<sub>2</sub> storage tank, as well as other direct and indirect costs. According to the IEAGHG [34], a new lime kiln cost approximately 19.8 M€ (inflation-adjusted). The OPEX included the catalyst renovation for methanation, the amine renovation, purchased electricity (77 €/MWh), required water, and operation and maintenance. The incomes were the savings in fossil natural gas (40 €/MWh), the synthetic natural gas sold (whenever surplus is found), the CO<sub>2</sub> taxes (84 €/tCO<sub>2</sub>), and the oxygen sold. The loan amortisation was set at 20 years, with 8,000 hours of operation per year [35]. The specific carbon capture costs, expressed in €/tCO<sub>2</sub> and €/tCaO, were described in Eqs. (24) and (25), respectively.

$$\text{Specific Carbon Avoidance Cost} = \frac{\left( \frac{\text{CAPEX}}{\text{Loan amortisation}} + \text{OPEX} - \text{Incomes} \right) 10^6}{\text{CO}_2 \text{ avoided} \times \text{Operating hours}} \left[ \frac{\text{€}}{\text{tCO}_2} \right] \quad (24)$$

**Table 4**  
Location and technical data of the PV solar field.

	Roof type 1	Roof type 1&2
<b>PV panels</b>		
Roof area (m <sup>2</sup> )	141,048	704,101
Solar area (m <sup>2</sup> )	98,734	380,621
Average system loss* (%)	35.6	35.6
Peak power (MWp)	14.8	56.8
Max. power production (MW)	11.5	44.1
Electricity production (MWh/year)	15,014	57,532
Equivalent operating hours (h)	1,013	1,013
<b>Li-ion battery</b>		
Round-trip efficiency (%)	85.0	85.0
Power capacity (MW)	0.74	2.84
Energy capacity (in time) (h)	4	4
Electricity processed (MWh/year)	603	2,405
Electricity provided (MWh/year)	512	2,044
Average charge (%)	9.5	10.1
<b>Electrolyser</b>		
Consumption (kWh/Nm <sup>3</sup> )	4.50	4.50
Power capacity (MW)	6.9	25.6
Nominal H <sub>2</sub> production (Nm <sup>3</sup> /h)	1,533	5,693
Nominal O <sub>2</sub> production (Nm <sup>3</sup> /h)	766	2,846
Usage of PV electricity (%)	92.5	91.3
Electricity processed (MWh/year)	13,893	52,524
H <sub>2</sub> production (Nm <sup>3</sup> /year)	3,087,288	11,671,965
O <sub>2</sub> production (Nm <sup>3</sup> /year)	1,543,644	5,835,983
Equivalent operating hours (h)	2,014	2,050
Average capacity factor (%)	23.0	23.4
Average H <sub>2</sub> production (Nm <sup>3</sup> /h)	352	1,332

\* Including effects related to the angle of incidence and temperature

$$\text{Specific Implementation Cost} = \frac{\left( \frac{\text{CAPEX}}{\text{Loan amortisation}} + \text{OPEX} - \text{Incomes} \right) 10^6}{\text{Lime production} \times \text{Operating hours}} \left[ \frac{\text{€}}{\text{tCaO}} \right] \quad (25)$$

## 4. Results and discussion

### 4.1. Renewable electricity and H<sub>2</sub> production

Following the methodology described above, the PV field was sized based on the available roof area. Then, the electrolyser was sized as a function of the load duration curve. Therefore, the results regarding the electricity and H<sub>2</sub> production were independent of the decarbonisation pathway selected for the lime kiln (only the size of the methanation plant had to be adjusted for each case). The Table 4 gathers the two PV scenarios analysed: (i) utilisation of the roof type 1, and (ii) utilisation of both the roof type 1&2. Type 3 roof was not considered due to the high investment required.

In PV systems, the ratio of solar area (usable area for power production) to available area (roof area) is typically 50-90%, depending on roof angle and surrounding shadows [51]. In our study, we assumed 70% for Type 1 and 50% for Type 2 (conservative approach). The total solar area for buildings ready for PV installation (i.e., Type 1) was 0.1 km<sup>2</sup>, equivalent to 15 MWp of PV capacity. When considering the roofs that required moderate rehabilitation or cleaning (i.e., Type 2), the total solar area increased to 0.38 km<sup>2</sup>, corresponding to 57 MWp of PV capacity. The auxiliary Li-ion batteries installed had round-trip energy storage capacities of 2.5 MWh (4 h × 0.74 MW × 0.85) and 9.7 MWh (4 h × 2.84 MW × 0.85), respectively.

The size of the electrolyser was chosen in order to maximise the energy that can be stored at nominal load,  $W_e^*$  (Fig. 8, left). Additionally, the criterion 2 described in Section 3.2 had to be applied, since there was a wide range of electrolyser sizes in which  $W_e^*$  was similar (e.g., for the roof Type 1&2, the value of  $W_e^*$  only varied ±1% for sizes between 19.8 MW and 25.6 MW, what can be observed in the flat-topped hill of the curve in Fig. 8, left). Hence, the selected electrolyser capacity was 6.9

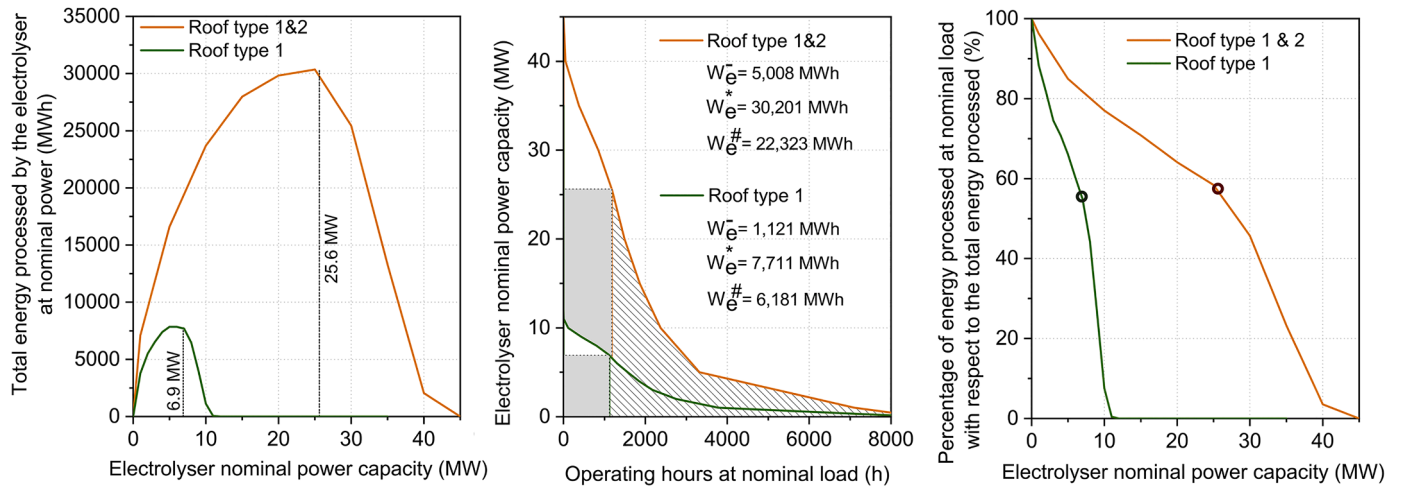


Fig. 8. Sizing of the electrolyser (6.9 MW for roof type 1, and 25.6 MW for roof type 1&2). Left: Total energy processed by the electrolyser at nominal power vs Electrolyser nominal power; Centre: Electrolyser nominal power vs Operating hours at nominal load (load duration curve); Right: Percentage of energy processed at nominal load with respect to the total energy processed vs Electrolyser nominal power.

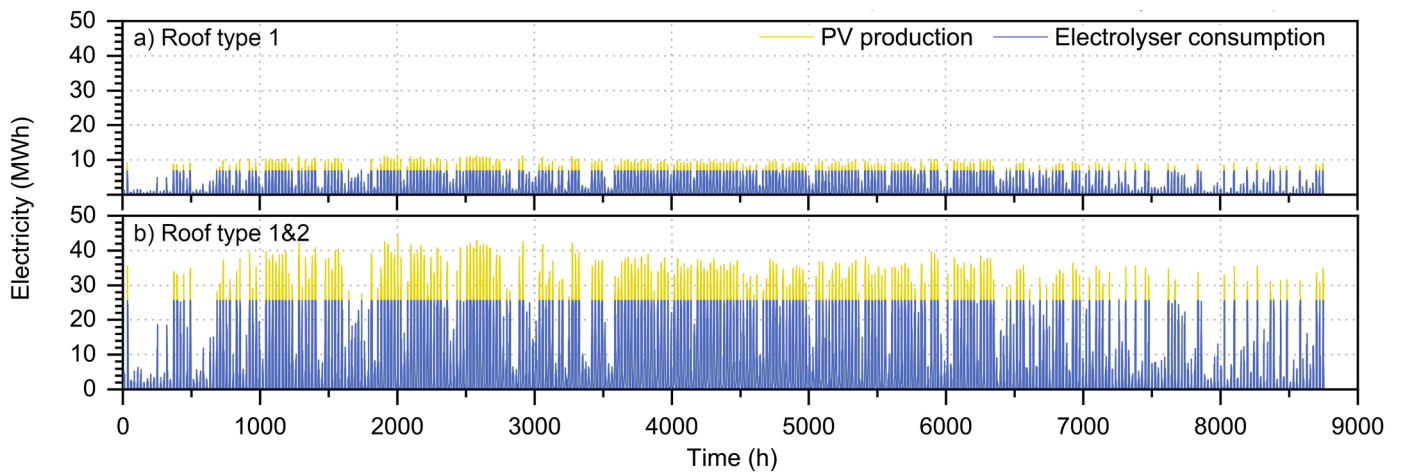


Fig. 9. PV electricity production (MWh) and Electrolysis electricity consumption (MWh) vs. time (h).

MW for the roof type 1 and 25.6 MW for the roof type 1&2. These solutions were depicted in the load duration curves of Fig. 8 (centre). The percentage of energy processed at nominal load relative to the total energy processed by the electrolyser, was between 55% and 57%. This point corresponds to a clear change in the slope of the curve of Fig. 8 (right), beyond which the percentage of energy stored at nominal load decreases faster (before this point, the percentage decreases just because the pattered area of the LDC increases, while beyond this point the percentage decreases faster because the grey area starts getting smaller).

With this installation size, the yearly electricity production of the PV field was 15.0 GWh for the roof type 1, and 57.5 GWh for the combination of roof type 1&2, both with 1013 equivalent operating hours (Table 4). The 91-92% of this electricity could be used in the electrolyser, see Fig. 9 (the rest corresponds to the blank area under the LDC in Fig. 8, which should be discarded or used in other parts of the iron-making plant). Throughout the year, the Li-ion battery processed 0.6 GWh and 2.4 GWh, respectively, resulting in the recovery of 0.5 GWh and 2.0 GWh of electricity after accounting for losses (yearly-average charge of 10%). Therefore, the Li-ion battery was not only useful to palliate cloud-coverage, but also for transferring some part of the sun production from the noon to the evening (if the Li-ion battery were not present, only the 87.6% of the PV electricity could have been used in the electrolyser, instead of the 91-92%). The yearly H<sub>2</sub> production was 3.1

and 11.7 million Nm<sup>3</sup>, for the roof type 1 and the roof type 1&2 scenarios, respectively. The electrolysis capacity factor was 23% (2014-2050 equivalent operating hours).

#### 4.2. CO<sub>2</sub> emissions and synthetic natural gas production

As shown in the previous section, the H<sub>2</sub> available for a given PV scenario was the same across all decarbonisation options, as it depended only on the installed PV capacity. Consequently, the methanation plant sizing, in terms of H<sub>2</sub> consumption, was the same for all decarbonisation options. As described in the methodology section, the smaller the methanation plant, the larger the H<sub>2</sub> tank needed to match the electrolyser's production with the methanation demand. However, small methanation sizes allowed higher capacity factors, which are desirable for steady operations. Therefore, we minimised the methanation plant size constrained by a maximum reasonable H<sub>2</sub> tank size of 66,740 Nm<sup>3</sup>. Thus, for Roof type 1, the nominal H<sub>2</sub> consumption of the methanation plant was 495 Nm<sup>3</sup>/h (1.5 MW<sub>H<sub>2</sub></sub>), and for Roof type 1&2 the nominal H<sub>2</sub> consumption was 2,140 Nm<sup>3</sup>/h (6.4 MW<sub>H<sub>2</sub></sub>). The equivalent operating hours were 6,238 h in the first case (71.2% capacity factor), and 5,454 h in the second case (62.3% capacity factor) (Fig. 10). Since the H<sub>2</sub> tank size was the same in both scenarios (Fig. 11), its contribution to maintaining high capacity factors in methanation are less significant as

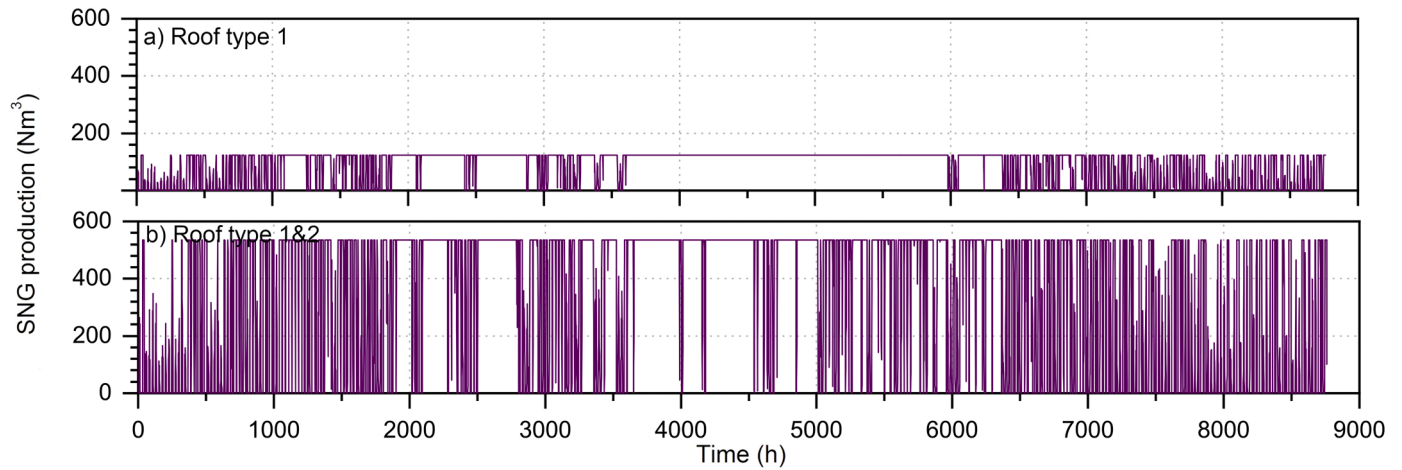


Fig. 10. Synthetic natural gas produced ( $\text{Nm}^3$ ) vs. time (h).

PV power increases. The methanation plant's efficiency was always around 82–83%. The synthetic natural gas produced differed slightly due to small differences in the flue gas composition entering the methanation process for each concept. In particular, this dependency was considered for through the parameters  $\epsilon_{mi}$  (gas conversion efficiency) and  $\nu_{mi}$  (stoichiometry of methanation) appearing in Eqs. (22) and (23).

Regarding  $\text{CO}_2$  emissions, the intrinsic characteristics of each concept led to different emission reductions (Table 5), as we had the option of providing heat for calcination with synthetic methane or  $\text{H}_2$ . Regarding the first PV scenario, in which only Roof type 1 was considered (Table 6),  $\text{CO}_2$  emissions reductions were either 2% or 22% with respect to a conventional air-blown lime kiln with no mitigation

measures. This difference mainly came from the strategy followed: the reduction was 2% if we partially replaced fossil natural gas by using the synthetic methane that was produced (carbon recycling), while the reduction was 22% if we avoided the fossil natural gas by directly using  $\text{H}_2$  (and selling the synthetic methane produced).

In the first case, we had the option of either amine scrubbing or oxy-fuel combustion to capture  $\text{CO}_2$  from the combustion. If we used amines, the energy penalty was 33.1 MJ/kg $\text{CO}_2$ , which was due to electrolysis consumption. If we used oxy-fuel combustion, the energy penalty was 51.1 MJ/kg $\text{CO}_2$ . The higher penalisation of the latter arose from the necessity of producing pure  $\text{O}_2$  with an air separation unit (380 kWh/t $\text{O}_2$  [52]), as the electrolyser could only provide the 10% of the  $\text{O}_2$  required in the kiln. In both cases, the natural gas used in the lime kiln was around

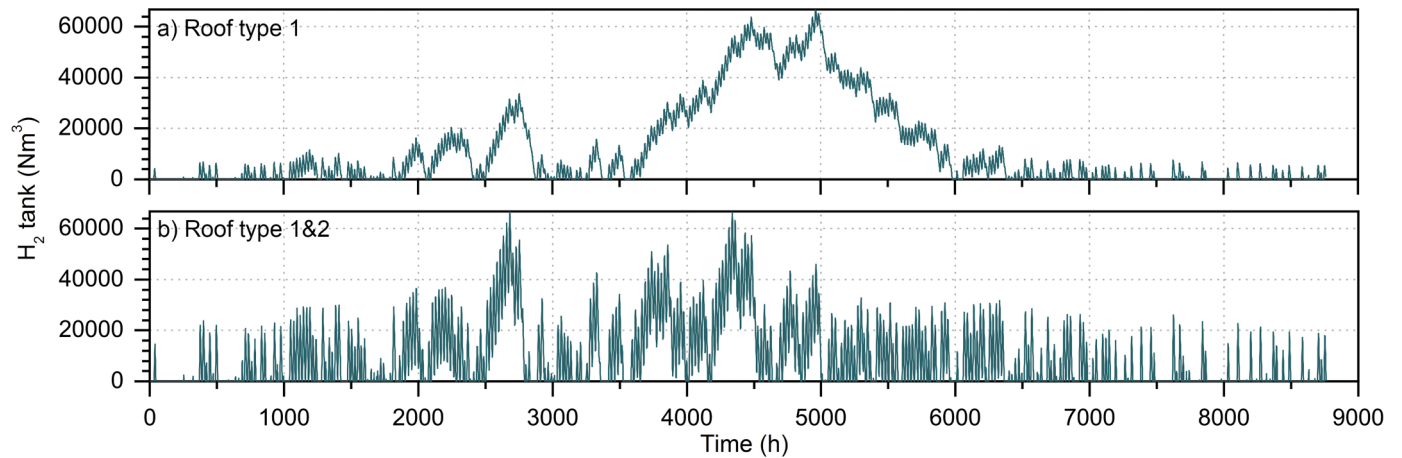


Fig. 11.  $\text{H}_2$  stored in the tank ( $\text{Nm}^3$ ) vs. time (h).

Table 5

$\text{CO}_2$  emissions, electricity consumption and specific energy penalty for the different decarbonisation pathways proposed. Minimum and maximum values depend on the total roof area for PV installation (corresponding to Roof type 1 and Roof type 1&2 areas, respectively).

Lime kiln technology	Air-blown	Air-blown + amines	Oxy-fuel	$\text{H}_2$ Oxy-fuel
$\text{CO}_2$ emissions (t $\text{CO}_2$ /year)	68,000	66,855 – 62,657	66,877 – 62,741	53,521 – 49,394
$\text{CO}_2$ reduction (t $\text{CO}_2$ /year)	-	1,510 – 5,707	1,488 – 5,624	14,844 – 18,971
$\text{CO}_2$ reduction (%)	-	2.2 – 8.3	2.2 – 8.2	21.7 – 27.7
Electricity consumption (TJ/year)				
Electrolyser ( $\text{H}_2$ for methanation)	-	50.0 – 189.1	50.0 – 189.1	50.0 – 189.1
Electrolyser ( $\text{H}_2$ for lime kiln)	-	-	-	348.8
ASU ( $\text{O}_2$ for lime kiln)	-	-	26.0 – 11.4	0.0
Total	-	50.0 – 189.1	76.0 – 200.5	398.8 – 537.9
Energy penalty (MJ/kg $\text{CO}_2$ )	-	33.1	51.1 – 35.6	26.9 – 28.3

Table 6

Technical data assessment for the different decarbonisation pathways proposed, using the roof area of Roof type 1 for PV installation.

Decarbonisation pathway	Air-blow + amines	Oxy-fuel	H <sub>2</sub> Oxy-fuel
<b>Lime kiln</b>			
Nominal production ( $t_{\text{lime}}/\text{h}$ )	8.3	8.3	8.3
Equivalent operating hours (h)	8410	8410	8410
Average capacity factor (%)	96.0	96.0	96.0
Yearly production ( $t_{\text{lime}}/\text{year}$ )	70,083	70,083	70,083
Natural gas yearly consumption ( $\text{Nm}^3/\text{year}$ )	6,659,038	6,659,038	-
From Power to Gas (%)	11.7	11.6	-
From gas grid (%)	88.3	88.4	-
H <sub>2</sub> yearly consumption ( $\text{Nm}^3/\text{year}$ )	-	-	21,529,600
O <sub>2</sub> yearly consumption ( $\text{Nm}^3/\text{year}$ )	-	14,825,232	11,967,430
Flue gas yearly production ( $\text{Nm}^3/\text{year}$ )	169,491,776	37,962,908	30,637,630
CO <sub>2</sub> yearly production ( $t_{\text{CO}_2}/\text{year}$ )	68,365	68,350	55,005
Recycled through Power to Gas (%)	2.2	2.2	2.7
Released to the atmosphere (%)	97.8	97.8	97.3
<b>Electrolyser (sized for PV panels)</b>			
Equivalent operating hours using PV (h)	2,014	2,014	2,014
Additional equivalent operating hours using grid to feed lime kiln (h)	-	-	6,386
Total equivalent operating hours (h)	2,014	2,014	8,400
Yearly H <sub>2</sub> production using PV ( $\text{Nm}^3/\text{year}$ )	3,087,288	3,087,288	3,087,288
Additional yearly H <sub>2</sub> production using grid ( $\text{Nm}^3/\text{year}$ )	-	-	9,789,186
Total yearly H <sub>2</sub> production ( $\text{Nm}^3/\text{year}$ )	3,087,288	3,087,288	12,876,474
<b>Additional electrolyser (required to fulfil lime kiln needs)</b>			
Power capacity (MW)	-	-	6.3
Equivalent operating hours (h)	-	-	8,400
Yearly H <sub>2</sub> production ( $\text{Nm}^3/\text{year}$ )	-	-	11,740,414
<b>Methanation</b>			
Conversion efficiency, $\epsilon$ ( $\text{Nm}^3_{\text{SNG}}/\text{Nm}^3_{\text{H}_2}$ )	0.2534	0.2498	0.2491
Stoichiometry, $\nu$ ( $\text{mol}_{\text{H}_2}/\text{mol}_{\text{CO}_2}$ )	4.0170	4.0766	4.0856
Nominal H <sub>2</sub> consumption ( $\text{Nm}^3/\text{h}$ )	495	495	495
Nominal SNG production ( $\text{Nm}^3/\text{h}$ )	125.4	123.6	123.3
Equivalent operating hours (h)	6,238	6,238	6,238
Average capacity factor (%)	71.2	71.2	71.2
Yearly SNG production ( $\text{Nm}^3/\text{year}$ )	782,068	771,016	768,856
Diverted to lime kiln (%)	100.0	100.0	0.0
Sold to the gas grid (%)	0.0	0.0	100.0
LHV <sub>SNG</sub> ( $\text{MJ}/\text{Nm}^3$ )	35.1	35.1	35.1
LHV <sub>SNG</sub> ( $\text{MJ}/\text{kg}$ )	49.5	49.5	49.6
Molar mass SNG ( $\text{kg}/\text{kmol}$ )	15.87	15.88	15.87
Efficiency LHV (%)	82.9	81.8	83.4
<b>H<sub>2</sub> tank</b>			
Maximum capacity ( $\text{Nm}^3$ )	66,740	66,740	66,740
Yearly average capacity (%)	17.7	17.7	17.7
<b>Other</b>			
Amine scrubbing plant size ( $t_{\text{CO}_2}/\text{h}$ )	0.24	-	-
O <sub>2</sub> production ( $\text{Nm}^3/\text{year}$ )	1,543,644	14,825,232	12,308,443
From ASU (%)	0.0	89.6	0.0
From Electrolyser (%)	100.0	10.4	12.5 + 39.8 + 47.7
O <sub>2</sub> for selling ( $\text{Nm}^3/\text{year}$ )	1,543,644	0	341,013

Table 7

Technical data assessment for the different decarbonisation pathways proposed, using the roof area of Roof type 1&amp;2 for PV installation.

Decarbonisation pathway	Air-blow + amines	Oxy-fuel	H <sub>2</sub> Oxy-fuel
<b>Lime kiln</b>			
Nominal production ( $t_{\text{lime}}/\text{h}$ )	8.3	8.3	8.3
Equivalent operating hours (h)	8410	8410	8410
Average capacity factor (%)	96	96.0	96.0
Yearly production ( $t_{\text{lime}}/\text{year}$ )	70,083	70,083	70,083
Natural gas yearly consumption ( $\text{Nm}^3/\text{year}$ )	6,659,038	6,659,038	-
From Power to Gas (%)	43.7	43.8	-
From gas grid (%)	56.3	56.2	-
H <sub>2</sub> yearly consumption ( $\text{Nm}^3/\text{year}$ )	-	-	21,529,600
O <sub>2</sub> yearly consumption ( $\text{Nm}^3/\text{year}$ )	-	14,825,232	11,967,430
Flue gas yearly production ( $\text{Nm}^3/\text{year}$ )	169,491,776	37,962,908	30,637,630
CO <sub>2</sub> yearly production ( $t_{\text{CO}_2}/\text{year}$ )	68,365	68,350	55,005
Recycled through Power to Gas (%)	8.4	8.2	10.2
Released to the atmosphere (%)	91.6	91.8	89.8
<b>Electrolyser (sized for PV panels)</b>			
Equivalent operating hours using PV (h)	2,050	2,050	2,050
Additional equivalent operating hours using grid to feed lime kiln (h)	-	-	3,782
Total equivalent operating hours (h)	2,050	2,050	5,832
Yearly H <sub>2</sub> production using PV ( $\text{Nm}^3/\text{year}$ )	11,670,650	11,670,650	11,670,650
Additional yearly H <sub>2</sub> production using grid ( $\text{Nm}^3/\text{year}$ )	-	-	21,529,600

(continued on next page)

Table 7 (continued)

Decarbonisation pathway	Air-blow + amines	Oxy-fuel	H <sub>2</sub> Oxy-fuel
Total yearly H <sub>2</sub> production (Nm <sup>3</sup> /year)	11,670,650	11,670,650	33,200,250
<b>Additional electrolyser (required to fulfill lime kiln needs)</b>			
Power capacity (MW)	-	-	-
Equivalent operating hours (h)	-	-	-
Yearly H <sub>2</sub> production (Nm <sup>3</sup> /year)	-	-	-
<b>Methanation</b>			
Conversion efficiency, $\epsilon$ (Nm <sup>3</sup> <sub>SNG</sub> /Nm <sup>3</sup> <sub>H<sub>2</sub></sub> )	0.2534	0.2498	0.2491
Stoichiometry, $\nu$ (mol <sub>H<sub>2</sub></sub> /mol <sub>CO<sub>2</sub></sub> )	4.0170	4.0766	4.0856
Nominal H <sub>2</sub> consumption (Nm <sup>3</sup> /h)	2,140	2,140	2,140
Nominal SNG production (Nm <sup>3</sup> /h)	542	535	533
Equivalent operating hours (h)	5,454	5,454	5,454
Average capacity factor (%)	62.3	62.3	62.3
Yearly SNG production (Nm <sup>3</sup> /year)	2,957,451	2,915,657	2,907,487
Diverted to lime kiln (%)	100.0	100.0	0.0
Sold to the gas grid (%)	0.0	0.0	100.0
LHV <sub>SNG</sub> (MJ/Nm <sup>3</sup> )	35.1	35.1	35.1
LHV <sub>SNG</sub> (MJ/kg)	49.5	49.5	49.6
Molar mass SNG (kg/kmol)	15.87	15.88	15.87
Efficiency LHV (%)	82.9	81.8	83.4
<b>H<sub>2</sub> tank</b>			
Maximum capacity (Nm <sup>3</sup> )	66,740	66,740	66,740
Yearly average capacity (%)	13.7	13.7	13.7
<b>Other</b>			
Amine scrubbing plant size (t <sub>CO<sub>2</sub></sub> /h)	1.1	-	-
O <sub>2</sub> production (Nm <sup>3</sup> /year)	5,835,982	14,825,232	16,600,125
From ASU (%)	0.0	60.6	0.0
From Electrolyser (%)	100.0	39.4	35.2 + 64.8 + 0.0
O <sub>2</sub> for selling (Nm <sup>3</sup> /year)	5,835,982	0	4,632,695

88% fossil and 12% renewable.

In the second strategy (22% CO<sub>2</sub> reduction) we used H<sub>2</sub> instead of fossil natural gas to provide the heat for calcination. This already provides a 20% reduction in CO<sub>2</sub> emissions compared to conventional operation, just from the total avoidance of the fossil fuels. The final emission reduction increases to 22% due to the production of synthetic methane with part of the flue gas.

Using H<sub>2</sub> as fuel, we needed to consume an additional 97 GWh/year of electricity to produce that hydrogen through electrolysis. Considering that the electrolysis capacity was 6.9 MW in this scenario, using our electrolyser to produce the additional H<sub>2</sub> resulted in increasing the electrolyser's operating hours from 2,014 to 16,057, which exceeds the annual period. Therefore, the H<sub>2</sub> lime kiln concept, under the Roof type 1 scenario, would require increasing electrolysis capacity to produce all the H<sub>2</sub> needed to replace the fossil natural gas, with the corresponding increase in investment. Assuming a maximum of 8400 operating hours (96% capacity factor), the total electrolyser capacity would be 13.2 MW,

rather than 6.9 MW. This additional operation would use grid electricity from renewable sources, ensuring that no further emissions are associated with this concept.

The energy penalty of the H<sub>2</sub> lime kiln, under the first scenario, was 26.9 MJ/kg<sub>CO<sub>2</sub></sub>. Because of the high H<sub>2</sub> production in this case, there is an excess of O<sub>2</sub> available for sale (487 t<sub>O<sub>2</sub></sub>/year). The total O<sub>2</sub> is obtained as a byproduct from the electrolyser, 12.5% originated from the PV electricity, 39.8% from the grid electricity when increasing the operating hours to 8400 h in the initial electrolyser (6.9 MW), and 47.7% from the grid electricity associated with the additional electrolysis capacity required (6.3 MW).

Regarding the second scenario, in which the Roof types 1&2 were considered (Table 7), the qualitative results were similar. Given the higher installed PV capacity, the CO<sub>2</sub> emission reduction was between 8% and 28%. Only one important difference must be highlighted: the H<sub>2</sub> lime kiln did not require an additional increase in the installed electrolysis capacity (25.6 MW), since 5,832 operating hours were sufficient to produce the H<sub>2</sub> for both the calcination and the methanation processes.

4.3. Cash flow and specific costs

CAPEX, OPEX, and Income vary greatly depending on the decarbonisation pathway and the roof type selected (Fig. 12). The CAPEX for the different pathways ranges from 99 M€ for air-blown + amines and roof type 1 to 391 M€ for both Oxy-fuel and H<sub>2</sub> Oxy-fuel, and roof type 1&2 (Table 8). In all cases, the electrolyser represents the largest CAPEX share, followed by the PV plant. The cases with roof type 1&2 have larger PV plants, which led to larger electrolyser sizes and therefore higher CAPEX. The amines cost is only considered in the air-blown + amines case, but it is relatively low, especially compared to the electrolyser cost. Methanation costs remain consistent across the three scenarios, with a notably lower magnitude compared than the electrolyser cost.

The OPEX varies from 3.0 M€/year for air-blown + amines and roof type 1 to 19.2 M€/year for H<sub>2</sub> Oxy-fuel and roof type 1&2, see Fig. 12. For the latter pathway, the biggest OPEX share is the electricity, for both roof types 1 and 1&2, due to the large amount of hydrogen required.

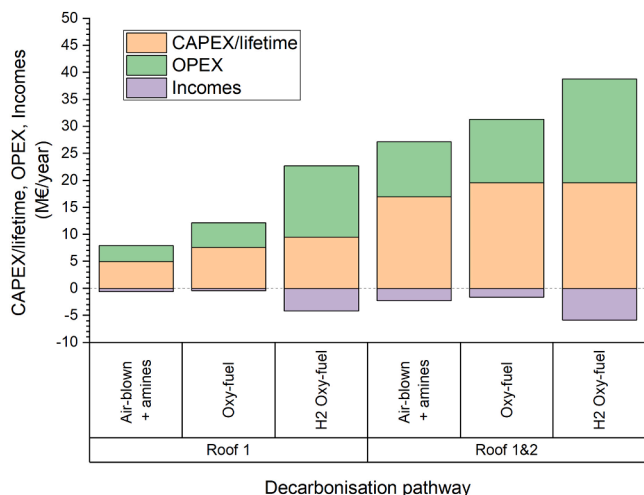


Fig. 12. Specific CAPEX (CAPEX/lifetime), OPEX and Incomes (M€/year), as a function of the decarbonisation pathway and the roof type.

**Table 8**  
Results of the economic analysis of the decarbonisation routes for lime kilns in ironmaking plants.

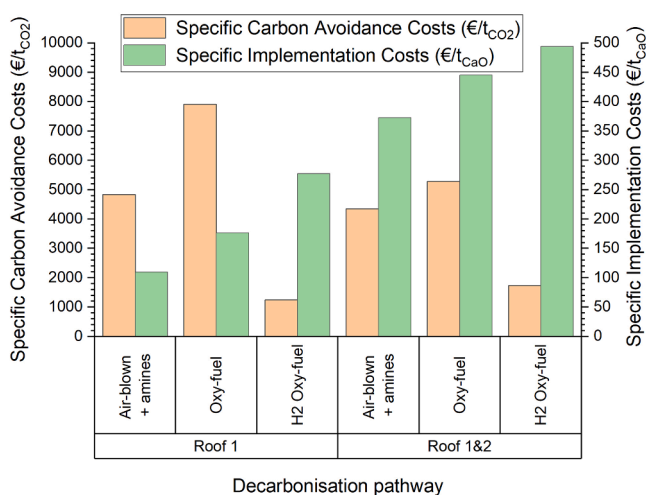
Scenario	Type 1 Air-blown + amines	Type 1 Oxy-fuel	Type 1 H <sub>2</sub> Oxy-fuel	Type 1&2 Air-blown + amines	Type 1&2 Oxy-fuel	Type 1&2 H <sub>2</sub> Oxy-fuel
<b>CAPEX [M€]</b>	98.9	152.3	190.0	339.1	391.1	391.1
Amine Plant	0.35	0.00	0.00	0.56	0.00	0.00
Electrolysis	12.1	12.1	23.1	44.8	44.8	44.8
H <sub>2</sub> storage						
H <sub>2</sub> storage tank	3.38	3.38	3.38	3.38	3.38	3.38
H <sub>2</sub> compressor	0.120	0.120	0.120	0.120	0.281	0.281
Methanation						
CO <sub>2</sub> compressor	0.016	0.016	0.016	0.039	0.039	0.038
Reactors	0.367	0.362	0.361	1.585	1.565	1.559
Catalyst	0.005	0.005	0.005	0.022	0.022	0.022
Lime kiln						
New oxy lime kiln	0.0	16	16	0.0	16	16
Solar						
PV plant	11.10	11.10	11.10	42.60	42.60	42.60
Batteries	1.48	1.48	1.48	5.68	5.68	5.68
Other direct costs						
Installation	11.27	17.37	21.67	38.67	44.60	44.60
Instrumentation/control	7.51	11.58	14.45	25.78	29.73	29.73
Piping	8.95	13.81	17.22	30.74	35.45	35.45
Electrical	2.89	4.45	5.56	9.92	11.44	11.44
Building	8.38	12.92	16.11	28.75	33.17	33.16
Indirect costs						
Engineering	9.24	14.25	17.78	31.73	36.60	36.59
Legal expenses	1.16	1.78	2.22	3.97	4.57	4.57
Construction expenses	9.82	15.14	18.89	33.71	38.88	38.88
Contingency	10.69	16.48	20.56	36.69	42.31	42.31
<b>OPEX [M€/year]</b>	2.97	4.57	13.18	10.19	11.75	19.21
Amine renovation	0.0006	0	0	0	0	0
Catalyst renovation	0.0008	0.0007	0.0007	0.0033	0.0032	0.0032
Electricity	0	0	7.47	0	0	7.46
Water	0.0041	0.0041	0.0078	0.0151	0.0151	0.0151
O&M	2.96	4.57	13.18	10.19	11.75	19.21
<b>Incomes [M€/year]</b>	0.61	0.43	4.19	2.29	1.61	5.85
Sold synthetic natural gas	0	0	0.30	0	0	1.13
Saved fossil natural gas	0.31	0.30	2.60	1.15	1.14	2.60
Oxygen	0.17	0	0.04	0.66	0	0.52
CO <sub>2</sub> taxes	0.13	0.13	1.25	0.48	0.47	1.60
<b>Specific costs [€/t<sub>CO2</sub>]</b>	4836	7906	1246	4356	5280	1735
<b>Specific costs [€/t<sub>CaO</sub>]</b>	110	176	277	373	445	494

Conversely, for the other pathways, the largest share is operation and maintenance (O&M). The Incomes ranges from 0.6 M€/y for oxy-fuel and roof type 1, to 5.9 M€/year for H<sub>2</sub> Oxy-fuel and roof type 1&2. In all cases, the main income is the natural gas (saved and sold) (Table 8). The income generated from the CO<sub>2</sub> taxes are significantly higher for the

H<sub>2</sub> Oxy-fuel cases. This divergence can be attributed to the electrolyser's extended operating hours. However, OPEX is always higher than the Income.

The specific carbon avoidance cost and the specific implementation cost, calculated with Eq. (24) and Eq. (25), are shown in Fig. 13. For roof type 1, the carbon avoidance cost is higher for Air-blown + amines and Oxy-fuel cases, owing to the lower operating hours of the electrolyser (2014 h), compared to the H<sub>2</sub> Oxy-fuel case (8400 h). For roof type 1&2, the same trend is observed, with higher costs for the two former cases (with 2050 h operating hours of the electrolyser), and lower costs for the latter case (with 5832 h). The implementation costs are directly related to the PV plant and electrolyser sizes, being higher in cases with roof type 1&2. Consequently, it is reasonable to assert that maximising the operating hours of the PtG system's electrolyser is recommended for optimal performance. Based on the results, H<sub>2</sub> Oxy-fuel is the best decarbonisation pathway for both roof type 1 and 1&2.

Implementing this technology after the current lime kiln's lifetime presents an opportunity to build a new one that meets new requirements, such as oxy-combustion with natural gas or with H<sub>2</sub>. Amortising the cost of the new lime kiln against the lime produced could reduce specific costs. Specifically, the oxy-fuel and H<sub>2</sub> oxy-fuel cases with roof types 1 and 1&2 pathways can reduce specific costs by 7% to 19%.



**Fig. 13.** Specific costs in €/t<sub>CO2</sub> and €/t<sub>CaO</sub>, as a function of the decarbonisation pathway and the roof type.

## 5. Conclusions

The iron and steel industry requires diverse decarbonisation strategies. Challenges remain in achieving complete decarbonisation, especially in essential processes like lime kilns. To address these challenges, innovative pathways utilising Power to Gas technology have been explored. By establishing carbon recycling concepts, these pathways aim to effectively reduce CO<sub>2</sub> emissions. The analysis includes various technologies: (1) conventional lime kiln integrated with amine scrubbing, (2) Oxy-fuel lime kiln, and (3) Oxy-fuel H<sub>2</sub> lime kiln. The integration of Power to Gas is facilitated through solar power, with PV panels installed on the rooftops of a steelworks located in Austria.

This study employs simulation to evaluate the integration of Power to Gas across different lime kiln technologies. The renewable resource has been managed through a decision-making methodology based on an hourly basis. This methodology utilises the load operating curve to maximise the energy processed by each piece of equipment at full load. Furthermore, the various process diagrams have been simulated in Aspen Plus under steady-state operation. Then, it has been evaluated economically using a 'factored estimated' methodology, which is based on the knowledge of major items of equipment (this methodology has an accuracy of  $\pm 30\%$ ).

The available roof area was determined by classifying roofs by their suitability for PV panel installation. Type 1 roofs are ready for PV panel installation and are not significantly affected by surrounding conditions like shadows or dust. Type 2 roofs require rehabilitation and/or moderate measures to mitigate ambient dust for PV panel installation, necessitating additional investment. Type 1 roofs alone accounted for 0.1 km<sup>2</sup>, equivalent to 15 MWp of PV capacity. Considering Type 2 roofs the total solar area increased to 0.38 km<sup>2</sup>, corresponding to 57 MWp of PV capacity. The selected electrolyser capacities were 6.9 MW for Type 1 roofs and 25.6 MW for Type 1&2 roofs. With these installation sizes, the annual electricity production for Type 1 roofs was 15.0 GWh, and for Type 1&2 roofs, it was 57.5 GWh, both with 1,013 equivalent operating hours. Approximately 91-92% of this electricity could be utilised in the electrolyser. The annual H<sub>2</sub> production was 3.1 million Nm<sup>3</sup> for Type 1 roofs and 11.7 million Nm<sup>3</sup> for Type 1&2 roofs, with an electrolysis capacity factor of 23%.

The methanation plant size was optimised while adhering to a maximum reasonable H<sub>2</sub> tank size of 66,740 Nm<sup>3</sup>. This resulted in a nominal hydrogen consumption of 495 Nm<sup>3</sup>/h for Roof type 1 and 2,140 Nm<sup>3</sup>/h for Roof types 1&2, with corresponding equivalent operating hours of 6,238 h (71.2% capacity factor) and 5,454 h (62.3% capacity factor), respectively. The methanation plant's efficiency consistently ranged between 82% and 83%.

In the first PV scenario involving only Roof type 1, CO<sub>2</sub> emissions reductions ranged from 2% to 22% compared to a conventional air-blown lime kiln with no mitigation measures. This variation depended on the strategy employed: a 2% reduction occurred when partially replacing fossil natural gas with synthetic methane, while a 22% reduction was achieved by directly using H<sub>2</sub> and selling the synthetic methane produced. Employing amine scrubbing or oxy-fuel combustion strategies for CO<sub>2</sub> capture resulted in energy penalties of 33.1 MJ/kg<sub>CO2</sub> and 51.1 MJ/kg<sub>CO2</sub>, respectively. The energy penalty of the H<sub>2</sub> lime kiln under the first scenario was 26.9 MJ/kg<sub>CO2</sub>. In the second scenario involving Roof types 1&2, CO<sub>2</sub> emissions were reduced by 8% to 28%, with conclusions similar to those in the first scenario.

Regarding the economic analysis, CAPEX ranges from 99 M€ to 391 M€, with the electrolyser representing the largest share. OPEX ranges from 3.0 M€/year to 19.2 M€/year, with electricity accounting for the largest share only for H<sub>2</sub> Oxy-fuel. Incomes range from 0.6 M€/year to 5.9 M€/year, mainly from natural gas savings. Carbon avoidance costs are higher for Air-blown + amines and Oxy-fuel due to lower electrolyser operating hours. Implementing H<sub>2</sub> Oxy-fuel is recommended against the other options, but all of them provide specific capture cost well beyond reasonable values (>1000 €/t<sub>CO2</sub>). Therefore, the complete

decarbonisation of the steel production chain remains a challenge, and it is far from achievable solely with H<sub>2</sub> and synthetic fuels.

## CRedit authorship contribution statement

**Manuel Bailera:** Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization. **Alexander García-Mariaca:** Writing – original draft, Validation, Methodology. **Cristian Barón:** Writing – original draft, Visualization, Validation, Methodology.

## 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.

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## Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.rineng.2026.110076](https://doi.org/10.1016/j.rineng.2026.110076).

## Data availability

No data was used for the research described in the article.

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