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Carbon-neutral cement: The role of green hydrogen

M. Bacatelo [a,*], F. Capucha [a], P. Ferrão [b], F. Margarido [b], J. Bordado [a,c]

[a] c5Lab – Sustainable Construction Materials Association, 2795-242, Linda-a-Velha, Portugal
[b] Center for Innovation, Technology and Policy Research – IN+, Instituto Superior Técnico, University of Lisbon, 1049-001, Lisboa, Portugal
[c] Centro de Recursos Naturais e Ambiente – CERENA, Av. Rovisco Pais 628, 1049-001, Lisboa, Portugal

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ABSTRACT

Business-as-usual (BAU) cement production is associated with a linear model that contributes significantly to global warming and is dependent on volatile energy markets. A novel circular model is proposed, by adding three power-to-gas system components to current production systems: a calcium-looping (CaL) CO_2 capture unit; water electrolysis for hydrogen and oxygen generation; and a methanation unit for synthetic natural gas (SNG) production. The paper presents the first analysis of the combined industrial-scale operation of these components in a closed loop, where the SNG fuels the cement kiln and the CaL unit, while the O_2 produced feeds it. The circular, hybrid, and BAU models are compared in three feasibility scenarios. It is concluded that the circular model outperforms the other alternatives environmentally, opening a potential pathway for the cement industry to achieve near net-zero CO_2 emissions, reduce energy dependence and improve economic efficiency.

1. Introduction

The current linear model of cement production causes negative environmental and economic impacts, resulting in increased greenhouse gas emissions, waste generation, and supply chain risks [1,2]. As a CO_2, energy, and material-intensive industry, the cement sector must urgently reduce its carbon footprint and dependence on volatile energy markets while meeting growing demand [3]. Despite the implementation of energy efficient techniques, alternative fuels and clinker-to-cement ratio reduction efforts, about 2/3 of the emissions remain unavoidable due to $CaCO_3$ calcination [4]. Carbon Capture and Utilization (CCU) offers a promising solution to reduce the CO_2 emissions and produce marketable CO_2-based fuels [5,6].

The Power-to-Gas (P2G) route is an appealing technological option due to the high demand for natural gas (NG) and its rising cost [7,8]. A P2G process can be based on the use of captured CO_2, including three main systems: (1) a CO_2 capture unit to capture CO_2 with an adequate purity level, (2) an electrolyser powered by renewable energy to produce green H_2 and (3) a methanation unit that generates synthetic natural gas (SNG), rich in methane (CH_4) through an exothermic reaction of H_2 with CO_2, forming CH_4 and H_2O, as described in reaction (R1) [9–12].

$$CO_2 + 4\,H_2 \rightarrow CH_4 + 2\,H_2O \qquad \Delta H = 165\ \text{kJ/mol} \tag{R1}$$

In the cement industry, oxyfuel combustion is known for its ability to produce a concentrated CO_2 flue gas by modifying the combustion atmosphere. This is achieved by burning fuel in pure oxygen instead of air, eliminating nitrogen at the combustion stage. As a result, the flue gas is mainly composed of CO_2 and water vapor. The condensation of water that follows produces a relatively pure CO_2 stream, making the capture of CO_2 simpler. However, implementing oxyfuel combustion requires modifications to the standard combustion process. In contrast, post-combustion capture techniques, which involve removing CO_2 from combustion flue gases, can be integrated into existing cement manufacturing facilities without extensive alterations [10,13–16]. Among these technologies, calcium-looping (CaL) post-combustion is a promising option for CO2 capture in cement kilns due to the (1) partial recovery of waste heat from the CO_2 capture unit; (2) cement industry's expertise in using CaO-bearing materials; (3) compatibility of CaO purge with cement raw meal and (4) minimal impact on the clinkering process [15–17]. A multi-criteria assessment of oxyfuel, and CaL and mono-ethanolamine (MEA) post-combustion considering environmental, technical, and economic indicators for the Portuguese cement industry, ranked CaL as the leading technology [18].

Four types of water electrolysis technologies produce green hydrogen: alkaline (AEL), anion exchange membrane (AEM), proton exchange membrane (PEM) and solid oxide electrolysis (SOE). These technologies differ in their electrolyte and operating conditions, but

* Corresponding author. c5Lab, Sustainable Construction Materials – Collaborative Laboratory Edifício Central Park, Rua Central Park 6, 2795-242, Linda-a-Velha, Portugal.

E-mail address: mbacatelo@c5lab.pt (M. Bacatelo).

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Abbreviations

AEL	Alkaline
AEM	Anion Exchange Membrane
AH	Yearly annual hours
AHP	Analytic Hierarchy Process
ASU	Air Separation Unit
ATIC	Portuguese cement industry association
BAU	Business-As-Usual
CaL	Calcium Looping
CAPEX	Capital Expenditures
CCU	Carbon Capture and Utilization
CEPCI	Chemical Engineering Plant Cost Index
CPU	Compression and Purification Unit
CR	Consistency Ratio
CT	Construct Time
EC	Electricity Consumption
EF	Emission Factor
EP	Electricity Production
ETS	Emissions Trading Systems
EU	European Union
FC	Fixed Capital Investment
i	Starting Year
IPCC	Intergovernmental Panel on Climate Change
IRR	Internal Rate of Return
KPIs	Key Performance Indicators
LCA	Life Cycle Assessment
m	Mass
MCDM	Multi-Criteria Decision-Making
MEA	Monoethanolamine
n	Index
NG	Natural Gas
NPV	Net Present Value
OL	Operational Lifetime
OPEX	Operational Expenditures
P_{inst}	Photovoltaic Installed Capacity
P2G	Power-to-Gas
PEM	Proton Exchange Membrane
PPA	Power Purchase Agreements
PV	Photovoltaic
r	Discount Rate
SNG	Synthetic Natural Gas
SOE	Solid Oxide Electrolysis
SPEC	Specific Primary Energy Consumption
SR	Stoichiometric Ratio
t	Time
TEA	Techno-Economic Assessment
TRL	Technology Readiness Level
WC	Working Capital
WSM	Weighted Sum Model
x	Scenario
y	Model

share similar operating principles, in which an electric current splits the bonds between H_2 and O in the water molecule, as indicated in reaction (R2) [19–21].

$$H_2O \rightarrow H_2 + \frac{1}{2}O_2 \quad \text{(R2)}$$

This paper proposes a novel circular model that integrates a realistic business-as-usual (BAU) cement model with P2G system components, including a CaL CO_2 capture unit, water electrolysis, and a methanation unit. Prior research has mainly focused on individual analyses of P2G system components, some of which have been applied to the cement industry [22–26]. In contrast, this paper provides the first comparative techno-economic and environmental analysis of the interconnected operation of these components on an industrial scale. Moreover, previous studies have overlooked the application of the oxygen by-product from water electrolysis and SNG use as fuel in the cement kiln, despite the potential for improved economic efficiency. This paper examines their prospective benefits. Notably, earlier investigations have mainly considered MEA CO_2 capture implemented on a small scale, further highlighting the novelty of this research in exploring these P2G systems on an industrial scale [6,10,27,28].

In this model, represented in Fig. 1, large-scale SNG is produced using cement-based CO_2 CaL captured emissions and on-site produced green hydrogen to create a new value-added chain through P2G. SNG fuels the cement kiln and the CaL CO_2 capture unit, in a loop, while the excessive fuel is fed into the existing NG grid. Additionally, the water electrolysis oxygen by-product is fed into the CO_2 capture unit, enhancing circularity.

2. Methodological framework

A multi-criteria decision-making (MCDM) methodological framework was developed for a techno-economic and environmental analysis of the circular cement model's implementation on a large scale. A "cradle-to-gate" boundary and a functional unit of 1 tonne of clinker were assumed. The MCDM methodology integrated the standardized techno-economic assessment (TEA) and life cycle assessment (LCA) methodology for CO_2 utilization detailed in Refs. [29,30].

The MCDM approach compared three models: a realistic BAU cement plant model based on primary data provided by the Portuguese cement industry association (ATIC), which compiled data from all the Portuguese cement plants in 2018, and two derived models, the circular model, and a hybrid one, with no SNG reuse or on-site H_2 production. The three models, described in detail in Section 2.1, are referred to as variable y in the equations.

For each model, three scenarios (worst, intermediate, and best-case) were compared to reflect distinct contexts of the industry's feasibility to implement the P2G route, represented by the variable x in the equations. The differences between the scenarios are summarized in the Supplementary Information.

The analysis considers the goals and preferences of the stakeholders, including Portuguese cement companies (Cimpor and Secil), in a transparent and consistent assessment of the techno-economic and environmental trade-offs of the models. The circular model aims to

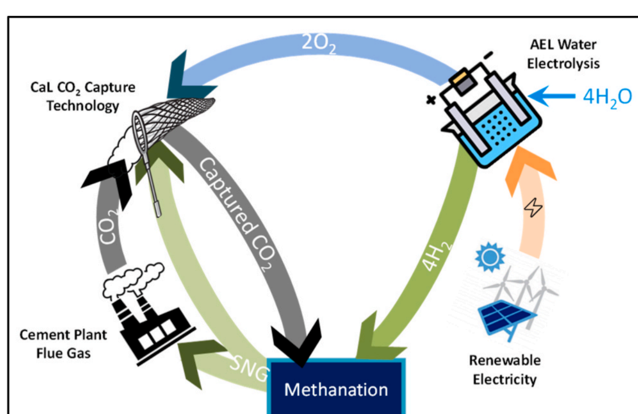

Fig. 1. Simplified proposed circular model.

address the problems in the cement industry mentioned in Section 1, by decreasing the dependence on a volatile energy market and reducing CO_2 emissions through a shift towards a circular economy, which matched the stakeholders' goals.

These goals were translated into 4 categories and 11 key performance indicators (KPIs), listed in Table 1, weighted and compared using the analytic hierarchy process (AHP) method. The weighted sum method (WSM) was applied to rank the three models in each scenario. These results can be used to design policy instruments and guide cement companies in defining their investment priorities, with the analysis adaptable to different stakeholder preferences by adjusting the weights assigned to the different KPIs. The simplicity and transparency of the WSM render it a suitable choice, as the decision-maker's preferences have a direct impact on the results [31].

To calculate these KPIs, the mass and energy balances were carried out for each model based on the BAU's primary data and secondary data built on top of it. The CaL CO_2 capture and methanation systems were based on the results of the simulation in Aspen Plus by Refs. [28,32], respectively, while the water electrolysis system was based on the model developed in HOMER open-source software [28].

These balances were used as inputs for the economic assessment of the models. It depends mainly on estimates of variable and fixable operational expenditures (OPEX), revenue and capital expenditure (CAPEX), which differ per model and scenario. The variable OPEX includes CO_2 costs and utility consumption, such as raw materials, fuels, electricity and water. The CAPEX was calculated considering the investment required to implement the P2G system components and the BAU CAPEX, which is the minimum amount of investment required to maintain current operations [33]. Cement, SNG and O_2 sales make up the revenue.

2.1. Models description

The different models analysed, represented in Fig. 2, consist of one or more of the following systems: "Clinker Production", "CaL CO_2 Capture Unit", "ASU", "Water Electrolysis" and "Methanation Unit". A production capacity of 1.08 $Mt_{clinker}$/year, a representative size for European cement plants, was assumed based on the average production of the three largest cement plants in Portugal (Cimpor-Alhandra, Secil-Outão and Cimpor-Souselas) [34–37].

Table 1
Key Performance Indicators per category. NA=Not applicable.

KPI Category	KPI	Units	Goal	Scenario dependent?
Economic	Net Present Value (NPV)	€/$t_{clinker}$	Maximize	Yes
	Internal Rate of Return (IRR)	%	Maximize	
	Payback Period	Years	Minimize	
Environmental	Total Net CO_2 Emissions	t_{CO2}/$t_{clinker}$	Minimize	Yes
	Total Net CO_2 Avoided	t_{CO2}/$t_{clinker}$	Maximize	
	Net Specific Primary Energy Consumption (SPEC)	kWh/$t_{clinker}$	Minimize	No
Technical	Direct CO_2 Converted	%	Maximize	Yes
	Technology Readiness Level (TRL)	NA	Maximize	No
	Energy independence	%	Maximize	
Eco-efficiency	Net CO_2 Avoided Costs	€/t_{CO2}	Minimize	Yes
	Net Cost of CO_2 Abatement		Minimize	

2.1.1. BAU model

The BAU cement plant model focuses on the energy-intensive and high CO_2-emitting "Clinker Production" stage. The raw materials (detailed in the Supplementary Information) are first mixed in different proportions to produce specific cement compositions. After grinding, the material enters a rotary kiln, passing through a pre-heater and pre-calciner, where it is gradually heated until calcination occurs at around 900 °C, releasing CO_2 from the calcium carbonate ($CaCO_3$). At temperatures up to 1450 °C, CaO reacts and agglomerates with silica, alumina and ferrous oxide to form clinker [34]. The "Clinker Production" system encompasses these processes.

2.1.2. Hybrid model

The hybrid model employs post-combustion CaL technology to capture the CO_2 emitted from the flue gas in the "Clinker Production" system. This technology is based on the reversible carbonation reaction and involves two interconnected circulating fluidized bed reactors (the carbonator and the calciner). Originally developed in Aspen Plus [32], this CaL configuration was adapted to a Portuguese cement plant [38]. The CaL calciner requires an oxidant, which is produced by the ASU and has a purity of 95%. Combustion temperature is controlled by mixing this oxidant with recycled combustion gases, resulting mainly in CO_2 and H_2O. After water condensation, a highly concentrated CO_2 stream is obtained [39].

The flue gas is directed to the carbonator, where CO_2 reacts with the CaO-based sorbent at around 650 °C under atmospheric pressure, forming $CaCO_3$. This calcium carbonate is transferred to the calciner, where the oxy-combustion of NG is carried out to reach a temperature of 950 °C (at atmospheric pressure), which is 30–50 °C above the calcination equilibrium temperature. This elevated temperature ensures complete calcination and sorbent regeneration, producing a CO_2-rich product stream with a dry molar purity of around 90%. The CaO-rich purge from the system is sent to the kiln and added to the raw meal. These processes are aggregated in the "CaL CO_2 Capture Unit" system, which generates substantial thermal energy via combustion in the calciner, recovered as high-temperature waste heat for electricity production [25,32,38,40].

The "Methanation Unit" uses a catalytic process for SNG production through CO_2 hydrogenation. It is based on the system developed in Aspen Plus [28], which includes a CO_2 compression and purification unit (CPU), an isothermal fixed bed reactor temperature-controlled by cooling water, and a purification system to meet the NG specifications. The CO_2-rich stream produced in the calciner requires further purification in a CPU due to excess oxidant and residual nitrogen and argon impurities from the ASU's oxygen stream. Consequently, the CO_2-rich stream is compressed up to 10 bar, resulting in a temperature increase to 250 °C [28].

The renewable H2 is assumed to be transported directly through a dedicated pipeline network, similar to the one proposed by the Rega Energy project [41]. The H_2 is first preheated to 280 °C, either in the reactor jacket or by heat exchange at the reactor outlet, and then mixed with the captured CO_2, considering a stoichiometric ratio of 4:1 [28]. The gas mix is fed to the methanation unit and further heated to 300 °C to produce raw SNG. The reaction mixture is further cooled down to 40 °C to remove water via condensation and reach a purity around 90 %. The produced SNG stream thus consists mainly of CH_4 and a smaller amount of H_2 (~7.5 v/v%), with some residual non-condensed moisture.

2.1.3. Circular model

The circular model has a few key modifications, when compared to the hybrid one. In particular, the "Water Electrolysis" produced both green H_2 and O_2, precluding the need for the "ASU". The analysis utilised AEL due to its proven scalability and established suitability for large-scale application and was based on the model developed in HOMER open-source software by Ref. [28]. While PEM electrolysers

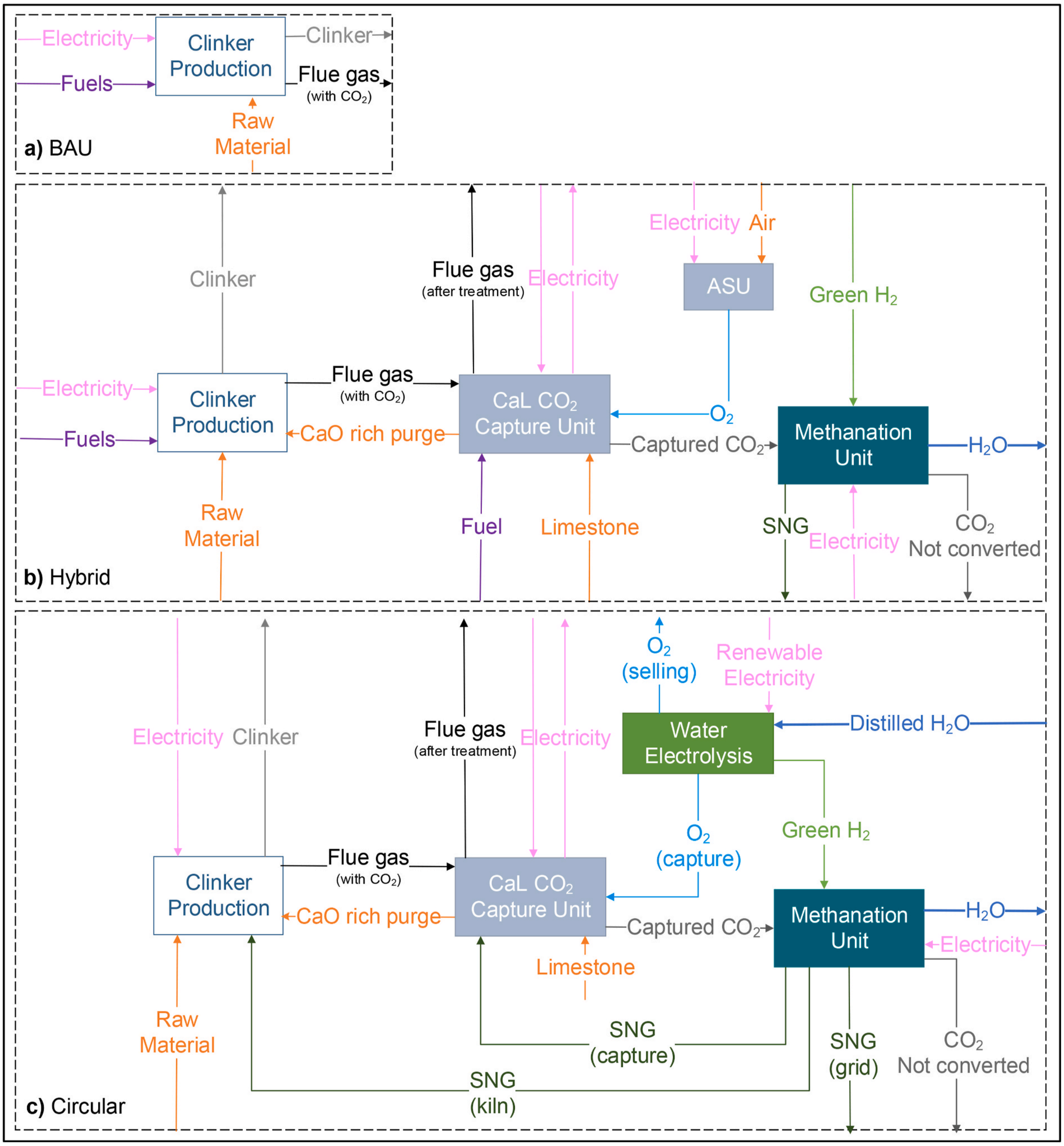

Fig. 2. Schematic overview of models to be compared.

exhibit high energy efficiency and current density, their large-scale deployment is still constrained by cost and durability challenges. Therefore, AEL is currently more suitable for widespread application [21,28,42–46].

The “Water Electrolysis” system operates at 70 °C and 10 bar, by utilizing a pair of electrodes immersed in an alkaline solution separated by a diaphragm, to split water into H_2 and O_2 by applying an electric current. The system operates at this specified pressure which bypasses the need for an additional compression stage for the hydrogen stream before its entry into the methanation unit [28]. The O_2, after compression in storage tanks (>99% purity at 10 bar), is partly fed to the CaL CO_2 capture unit while the surplus is sold to the market [21].

The stoichiometric amount of H_2 production via water electrolysis is around 9 kg_{H2O}/kg_{H2} [47]. However, considering the process efficiency, water purity, water loss from the system through periodic hydrogen purge and that both H_2 and O_2 leave the electrolyser wet, a water consumption of 10 kg_{H2O}/kg_{H2} was assumed [47–49]. The required renewable electricity is supplied by a 10 MW photovoltaic system (PV), slated for installation in Cimpor’s cement plants by 2025, and green power purchase agreements (PPA) [50]. In the circular model, the SNG produced in the “Methanation Unit” is recycled back to the “Clinker Production” and “CaL CO_2 Capture Unit” systems, replacing the fuels used in the other models for these systems, while the remaining SNG is injected into the grid.

In the circular system, the calcium-looping CO_2 capture process exhibits a capture efficiency of 92%, the electrolyser operates with an energy efficiency of 68%, and the methanation process achieves a conversion rate of 92% of CO_2 to CH_4, underpinning the system’s potential for circularity.

2.2. Key performance indicators

The economic evaluation of the models is based on a discounted cash flow approach. The main assumptions are summarized in the Supplementary Information.

2.2.1. Economic analysis

The economic viability of the models can be assessed by analysing their financial flows, using various KPIs. The NPV KPI sums the discounted future cash flows of each model at a given discount rate, as shown in equation (1) [51,52]. IRR is the interest rate that equates the NPV to zero [53,54]. The payback period is the time, in years, for net cash inflows to recover the initial investment [55].

$$NPV = \sum_{t=i}^{CT+OL} \frac{(Revenue + OPEX)_{t,x,y}}{(1+r)^t} + \sum_{t=i}^{CT} \frac{CAPEX_{t,x,y}}{(1+r)^t} \quad (1)$$

where:

- t = time.
- i = Starting year (2028).
- CT = Construction time (2028–2030).
- OL = Operational lifetime (2031–2055).
- r = Discount rate (8%) [23].
- OPEX and CAPEX are negative.
- x = Scenario.
- y = Model.

2.2.2. Environmental analysis

To evaluate the environmental impact of the different models, three KPIs were calculated. The "Total Net CO_2 Emissions" KPI, in $t_{CO2}/t_{clinker}$, considers the CO_2 converted into SNG, the direct CO_2 emissions from the calcination reaction and fuel consumption (fossil, alternative, biomass and SNG) and the indirect CO_2 emissions from the electricity consumption (EC), as indicated in equation (2). Renewable electricity and H_2 consumption were assumed to generate 0 emissions.

$$(Total\_Net\_CO_2Emissions)_{x,y} = (CO_2Calcination)_x + (CO_2Fuels)_{x,y} + (CO_2EC)_y - (CO_2Converted)_y \quad (2)$$

Two approaches were used to calculate the "Total Net CO_2 Emissions" of the models. The "Literature" method, based on secondary data, followed the IPCC Guidelines for estimating the CO_2 emissions from calcination. CO_2 emissions from fuel combustion were calculated as the product of fuel consumption and its emission factor (EF), see equation (3) [56]. The "Real/Adjusted" method, used primary data provided by ATIC on the BAU CO_2 content in the flue gas. The minimum and maximum values between the methods were reflected in the best and worst-case scenarios, respectively, while the intermediate scenario used the average of both methods, for each CO_2 category.

$$(CO_2Fuels)_{x,y} = (Energy\_Consumption)_y \times (Fuels\_EF)_{x,y} \quad (3)$$

Indirect CO_2 emissions from electricity were calculated based on both primary (EC for clinker production) and secondary data (electricity consumption and production — EP — in the new systems) and the Portuguese electricity emission factor, as indicated in equation (4).

$$(CO_2EC)_y = (Grid\_EC - CaL\_EP)_y \times Electricity\_EF \quad (4)$$

The "CO_2 converted" is defined as the CO_2 emissions that were converted into CH_4 in the methanation reaction, which depend on both the capture and methanation efficiencies (η), as expressed in equation (5).

$$(CO_2Converted)_{x,y} = (CO_2Calcination + CO_2Fuels)_{x,y} \times \eta_{CO_2Capture} \times \eta_{Methanation} \quad (5)$$

The "Total Net CO_2 Avoided" KPI measures the total net CO_2 emissions avoided in the cement plant due to the integration of model y compared to the BAU, as indicated in equation (6).

$$(Total\_Net\_CO_2Avoided)_{x,y} = (Total\_Net\_CO_2Emissions)_{x,y=BAU} - (Total\_Net\_CO_2Emissions)_{x,y} \quad (6)$$

Finally, the net SPEC, in kWh/$t_{clinker}$, was calculated considering the net primary energy consumption from the fuel combustion (including the SNG and green H_2), grid and renewable electricity (both on-site and from PPA) and the EP in the "CaL CO_2 capture Unit", as shown in equation (7).

$$(Net\_SPEC)_y = (Fuels\_Consumption)_y + (Renewable\_EC)_y + (Grid\_EC)_y - (CaL\_EP)_y \quad (7)$$

2.2.3. Technical analysis

The technical assessment of the different models relied on three KPIs. The "Direct CO_2 Converted", calculated using equation (8), measures the ratio between the CO_2 converted into SNG and the direct CO_2 emissions resulting from the calcination reaction and the fuel consumption.

$$(Direct\_CO_2Converted)_{x,y} = \frac{(CO_2Converted)_y}{(CO_2Calcination)_x + (CO_2Fuels)_{x,y}} \quad (8)$$

The "Energy Independence" KPI, presented in equation (9), measures the cement plant's independence from energy prices and its supply safety [57] through the ratio between the on-site electricity production (from the CaL and solar PV) and the SNG produced with the "Net SPEC".

$$(Energy\_Independance)_y = \frac{(CaL\_EP + PV\_EP)_y + (Produced\_SNG)_y}{(Net\_SPEC)_y} \quad (9)$$

The TRL scale is used to define the technological maturity of the overall model being analysed, and it is equal to the lowest TRL of its constituent process units, measured between 1 (basic principles observed) and 9 (commercial operation in relevant environment) [27,58].

2.2.4. Eco-efficiency analysis

Two eco-efficiency indicators, related to the circular economy goal of producing more whilst extracting fewer resources, were considered [30]. The "Net CO_2 Avoided Cost", indicated in equation (10), in €/$t_{CO2\ avoided}$, is defined as the quotient between the total additional costs of model y compared to the BAU, in €/$t_{clinker}$, and the total net CO_2 avoided, in $t_{CO2}/t_{clinker}$.

$$[Net\_CO_2Avoided\_Cost]_{x,y} = \frac{(Total\_Costs)_{x,y} - (Total\_Costs)_{x,y=BAU}}{(Total\_Net\_CO_2Avoided)_{x,y}} \quad (10)$$

The "Net Cost of CO_2 Abatement", in equation (11), is a similar concept; however, it accounts for the added revenue of model y compared to the BAU. This value is negative when the revenue of model y is higher than its costs and this difference is superior to the one verified in the BAU.

$$[Net\_Cost\_CO_2Abatement]_{x,y} = \frac{(Total\_Costs - Revenue)_{x,y} - (Total\_Costs - Revenue)_{x,y=BAU}}{(Total\_Net\_CO_2Avoided)_{x,y}} \quad (11)$$

For both eco-efficiency KPIs, the CO_2 price was assumed in the BAU model, as by definition it would result in a mathematical indeterminate form of 0/0.

3. Mass and energy balances

Mass and energy balances for each system were calculated from its inputs and outputs. This section presents the main results, while the Supplementary Information contains important numerical data such as key economic and technical assumptions, detailed mass and energy balances, and intermediary calculations. These details are crucial for a thorough understanding and validation of the research findings. Equation (12) describes the mass calculation of H_2, CH_4 and O_2 based on the stoichiometric ratio of reactions (R1) and (R2) using direct CO_2 as limiting reagent.

$$m(\text{unknown}) = SR \times \frac{m(\text{known})}{MM\ (\text{known})} \times MM(\text{unknown}) \tag{12}$$

where:

- $m(\text{unknown})$ = unknown mass of H_2, CH_4 or O_2.
- SR = Stoichiometric ratio of $CH_4/CO_2 = 1$, $H_2/CO_2 = 4$, or $O_2/H_2 = 0.5$.
- MM (known) = molecular mass of the known quantity of $CO_2 = 44.01$ or $H_2 = 2.02$ [g/mol].
- MM (unknown) = molecular mass of the unknown quantity of H_2, $CH_4 = 16.05$ or $O_2 = 32$ [g/mol].

3.1. Clinker production

The clinker production system requires electricity, fuels and raw materials as inputs. These values were based on primary data provided by ATIC. Clinker production is energy-intensive, requiring 1041 kWh of heat to produce one tonne. Fossil fuels (petcock and fuel oil) make up around 60% of the fuels used, while 36% are alternative waste derived fuels and the remaining 4% biomass (wt. %). To produce one clinker tonne, 1.42 tonnes of raw materials are required, with 97.3% being primary and 2.7% are alternative (wt.%). The introduction of the CaO rich purge from the CaL CO_2 capture system reduces raw material consumption by 2.46% (1.39 $t/t_{clinker}$) in the hybrid and circular models [23]. Additionally, a significant amount of electricity (114 $kWh/t_{clinker}$) is required. The main outputs of this system are the clinker (one tonne) and the flue gas, which contains, among other gases, 0.82 t_{CO2} (BAU and hybrid models) or 0.73 t_{CO2} (circular model).

3.2. Air separation unit

In the hybrid model, the ASU consumes 216 kWh to produce 0.44 tonnes of oxygen required to feed the CaL CO_2 capture system per clinker tonne [23].

3.3. CaL CO_2 capture

The "CaL CO_2 capture Unit" in the hybrid and circular models has five inputs (fuels, electricity, O_2, flue gas and limestone) to produce two outputs (captured CO_2 and electricity), which were based on secondary data [23]. This process consumes 1072 kWh of NG (hybrid model) or SNG (circular model) per clinker tonne. The electricity demand is 65.6 $kWh/t_{clinker}$, however, the high operating temperatures allow the recovery of heat introduced with the fuel in the calciner, which can be used to produce 740 $kWh/t_{clinker}$ in a Rankine cycle for sorbent regeneration. The CO_2 from the flue gas of "Clinker Production" and that formed in the calciner by fuel combustion is captured with a 94% CO_2 capture ratio, with the remaining 6% being released into the atmosphere. Therefore, the captured CO_2 corresponds to 0.98 $t_{CO2}/t_{clinker}$ in the hybrid model, and 0.89 t_{CO2} in the circular model. The process also requires an oxygen input of 0.44 $t_{O2}/t_{clinker}$ and a $CaCO_3$ makeup of 49 $kg_{CaCO3}/t_{clinker}$ for both models [25,32,38].

3.4. Methanation

The "Methanation Unit" required for the hybrid and circular models uses captured CO_2 from the "CaL CO_2 capture unit", green H_2 and electricity to produce the SNG main output. The necessary H_2 was estimated to enter the process at the SR of H_2:$CO_{2(captured)}$= 4:1 [28]. The produced SNG was based on the SR of $CO_{2(captured)}$:CH_4 = 1:1 and assuming a 92% reactor CO_2 conversion, using a Ni/CeO_2 catalyst [28, 59]. In the hybrid model, the total SNG produced (4780 $kWh/t_{clinker}$) is fed into the existing NG grid while in the circular model part of it is recirculated into the "Clinker Production" system (1041 $kWh/t_{clinker}$) and the "CaL CO_2 Capture Unit" (1072 $kWh/t_{clinker}$). This system has an electricity demand of 0.03 $MWh/t_{clinker}$ (assuming 1.95 kWh/GJ_{SNG} of electricity consumption [6]).

3.5. Water electrolysis

In the circular model, the "Water Electrolysis" system requires renewable energy and distilled water inputs to produce green H_2 and O_2 outputs. The oxygen is separated into two streams: the O_2 (capture) is fed to the CaL CO_2 capture unit at 0.44 $t_{O2}/t_{clinker}$ while the excess O_2 (selling) is sold to the market at 0.86 $t_{O2}/t_{clinker}$ [21]. The water input required, considering a mass ratio of 10 kg_{H2O}/kg_{H2} [49], is 1.64 $t_{H2O}/t_{clinker}$. The renewable electricity demand of 9.57 $MWh/t_{clinker}$ (assuming 58.27 MWh/t_{H2} of electricity consumption [60]), is provided by a PPA (9.54 $MWh/t_{clinker}$) and an on-site PV system (0.02 $MWh/t_{clinker}$ — see equation (13). Cimpor aims to increase energy independence by integrating PV, which is part of a forward-looking strategy to incorporate sustainable energy solutions. This reflects their plans to install PV systems across their cement plants by 2025 [50].

$$\text{Electricity\_On\_Site\_PV} = \frac{P_{inst}[MW] \times a \times AH\left[\frac{h}{year}\right]}{m_{clinker}\left[\frac{t_{clinker}}{year}\right]} \tag{13}$$

where:

- P_{inst} = PV installed capacity (10 MW) [50].
- a = Capacity factor in Portugal (0.27) [61].
- AH = Yearly annual hours (8760 h/year) [61].
- $m_{clinker}$ = Annual clinker production (1,082,598 $t_{clinker}$/year) [ATIC].

4. Economic assessment

To assess the models' techno-economic performance, their CAPEX, OPEX and revenue were calculated. Economic data was reported using 2021 prices and adjusted through the Chemical Engineering Plant Cost Index (CEPCI), as indicated in equation (14), when not directly available [62].

$$C = C_0 \times \left(\frac{CEPCI}{CEPCI_0}\right) \tag{14}$$

where:

- C = Cost in 2021 [€].
- C_0 = Base cost [€].
- CEPCI = CEPCI in 2021 (708.0) [63].
- $CEPCI_0$ = Base CEPCI.

The economic calculations differ across the three scenarios, as

Table 2
Parameters considered for each scenario.

Economic Type	Parameter/Scenario	Worst	Intermediate	Best	Explanation/Reference
Variable OPEX	General utilities and consumables prices	Medium + 50%	Medium	Medium - 50%	Supplementary Information
	CO_2 Price [€/t]	58.83	90.32	121.03	Calculated from [64–66]
	Purchased green H_2 in hybrid model [€/kg]	4	2.5	1.5	Based on the graphical information provided by Ref. [67] filtered for "end use application/Industrial feedstock"
CAPEX	BAU	Medium + Standard error	Medium	Medium–Standard error	Table 4
	CaL CO_2 Capture ASU Electrolysis O_2 Compression and Storage	Medium + 35%	Medium	Medium - 15%	
Revenue	Cement Price [€/t]	122	133	152	Adapted from [68]
	SNG Price [€/GJ]	4.97	9.94	14.91	Market price of NG in Portugal (2021) [69]
	Oxygen price [€/kg]	1	4	7	The oxygen is priced at varying rates of 1, 4, or 7 €/kg depending on the scenario, and is suitable for both industrial and medical purposes. It is worth noting that medical oxygen has a higher market value [70]

indicated in Table 2.

4.1. CAPEX

The BAU CAPEX was 8.43 ± 2.66 €/$t_{clinker}$, which is the average value of Secil's reported annual CAPEX between 2013 and 2018 (adjusted to €2021), normalized using the annual national clinker production, and the corresponding standard error [71–75]. The CAPEX of the new systems includes the fixed capital investment (FC) and working capital (WC). FC accounts for the total capital required to supply the manufacturing and plant facilities, including three cost types: direct (e.g. equipment purchase and installation, piping and electrical systems, instrumentation, buildings and service facilities), indirect (such as legal and construction expenses, engineering and supervision costs, contractor's fees and contingencies) and other [6,62]. WC represents the capital needed for plant operation, typically between 10 and 20 % of the FC, with a value of 15% assumed [62].

Table 3 presents the annual CAPEX allocation for the cement plant, following the guidelines in Ref. [76]. The estimated accuracy of the new systems' CAPEX is +35%/-15%, reflecting the worst and best-case scenarios, respectively [77].

The CAPEX of the new systems was calculated by the six-tenths factor rule, described in equation (15), which compares the capacity and investment of the studied process to those of a similar installation in the cement industry, with an appropriate index of 0.6 [62].

$$\text{New\_System\_CAPEX} = \text{New\_Systems\_CAPEX}_0 \times \left(\frac{A}{A_0}\right)^n \tag{15}$$

where:

- $\text{New\_Systems\_CAPEX}_0$ = Reference CAPEX of the new system obtained from literature.
- A = Capacity of the cement plant.
- A_0 = Capacity of the reference cement plant.
- n = index (0.6).

Table 3
CAPEX allocation [76].

CAPEX allocation	Time	Year
$(\text{BAU\_CAPEX})_x \times (\text{OL} + \text{CT}) + 40\%(\text{New\_Systems\_CAPEX})_{x,y} \times \text{OL}$	1	2028
$30\%(\text{New\_Systems\_CAPEX})_{x,y} \times \text{OL}$	2	2029
$30\%(\text{New\_Systems\_CAPEX})_{x,y} \times \text{OL}$	3	2030

Table 4 displays the normalized CAPEX, in €/$t_{clinker}$, based on the calculated CAPEX of the new systems, provided in the Supplementary Information. Notably, the additional expenses incurred in the commercialization of oxygen, such as logistics, distribution, and transportation, are not factored into the analysis.

4.2. OPEX

OPEX comprises both variable and fixed costs. The variable costs encompass CO_2 production, raw materials, electricity and fuel consumption expenses while fixed costs are associated with operation and maintenance (equivalent to 7% of the CAPEX [5]). The present analysis excludes the workers, supervisors, laboratory work and patents costs. The variable OPEX was determined by calculating the CO_2 costs and the sum of the product of each consumable's price and its quantity consumed for every model and scenario.

The CO_2 costs were assumed to be 60% of the European Union (EU) Emissions Trading System (ETS) in 2026, increasing linearly to 100% by 2030 onwards for the intermediate and best-case scenarios or by 2035 for the worst scenario [64,65]. The EU-ETS values were based on projections for 2020, 2030 and 2050, including minimum, average and maximum prices [66]. A linear interpolation method was used to estimate values between 2028 and 2050 and extrapolated to other years. The estimated average CO_2 price was 58.83 €/t_{CO2}, 90.32 €/t_{CO2} and 121.03 €/t_{CO2}, for the worst, intermediate and best-case scenarios, respectively. Note that the current EU carbon permit (as of March 2023) is 100.74 €/t_{CO2}, which falls between the intermediate and best-case scenarios [78].

5. Results and discussion

The present section is divided into five sub-sections, corresponding to the 4 KPIs category results and the MCDM interpretation.

5.1. Economic KPIs

Fig. 3 depicts the total costs, which are the sum of the normalized CAPEX and the fixed and variable OPEX. Fixed OPEX represents a minor fraction of the total costs (<1%), while variable OPEX, which includes CO_2 costs, utilities, and consumables, is the largest cost contributor (>91%) in both the BAU and hybrid models. However, the circular model incurs significantly higher CAPEX costs (41%–54% of total costs) than other models. Accordingly, the circular model has the highest total costs, followed by the hybrid model and, lastly, the BAU. Decreases in CAPEX, utilities and consumables (mainly electricity and fuel), explain the cost reduction from the worst to the best scenario in each model,

Table 4
Normalized CAPEX per model and scenario [€/$t_{clinker}$].

Scenario	Worst			Intermediate			Best		
System/Model	BAU	Hybrid	Circular	BAU	Hybrid	Circular	BAU	Hybrid	Circular
BAU	11	11	11	8	8	8	6	6	6
CO_2 Capture	0	14	14	0	10	10	0	9	9
ASU	0	4	0	0	3	0	0	3	0
Electrolysis	0	0	404	0	0	299	0	0	254
O_2 Compression and Storage	0	0	0.07	0	0	0.05	0	0	0.04
Methanation	0	7	7	0	5	5	0	4	4
Total CAPEX [€/$t_{clinker}$]	11	36	435	8	27	323	6	21	273

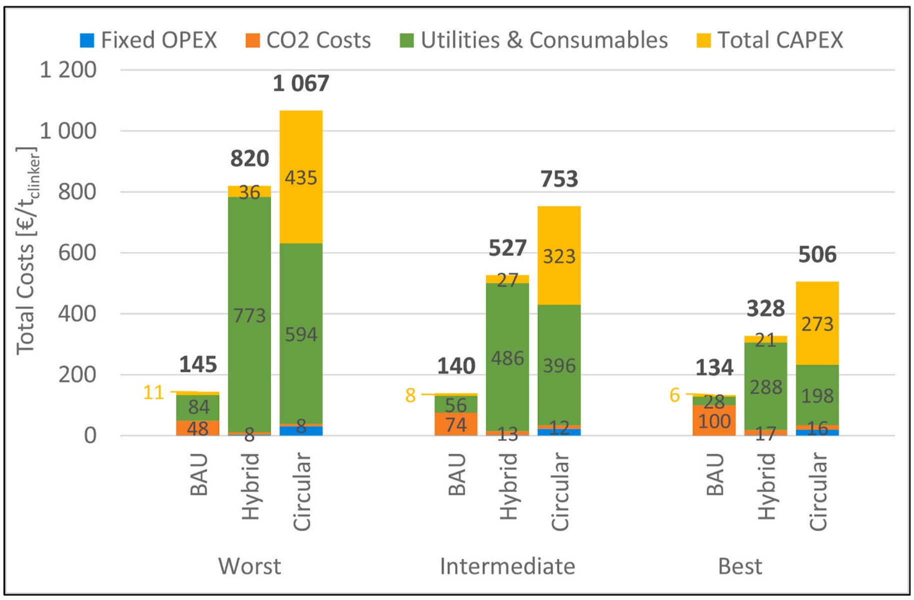

Fig. 3. Clinker production's total costs [€/$t_{clinker}$].

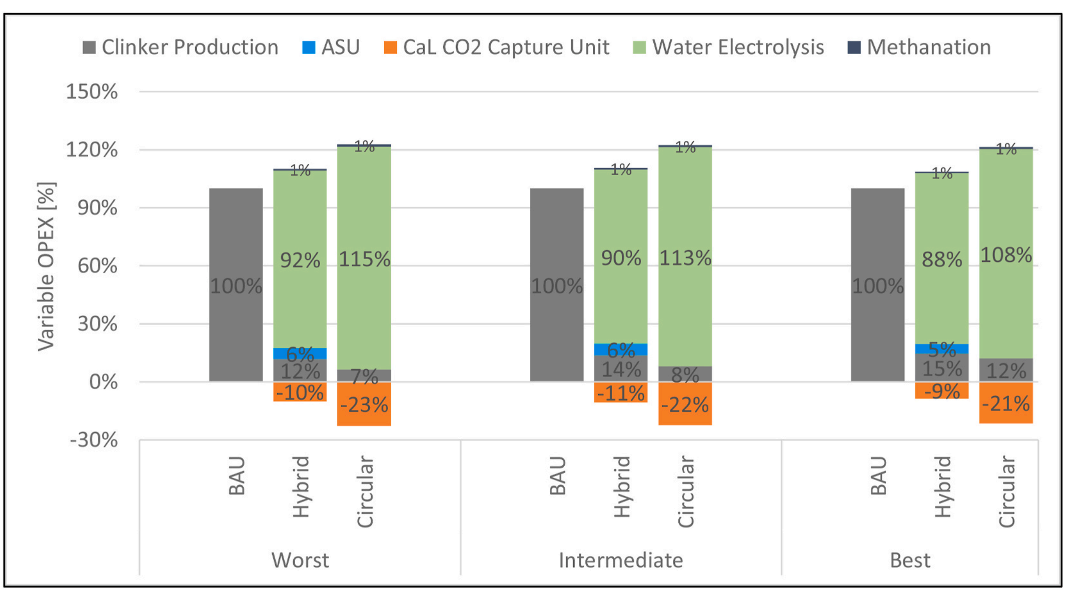

Fig. 4. System's contribution to variable OPEX [%].

despite the increased CO_2 costs.

The impact of each system in the variable OPEX is detailed in Fig. 4. The "Water Electrolysis" system has the highest impact in both the hybrid and circular models due to the high cost of purchased green H_2 and the electrolysis' required electricity. Conversely, the "CaL CO_2 capture" system negatively impacts the variable OPEX due to the electricity production.

The revenue of each model, displayed in Fig. 5 was calculated as the

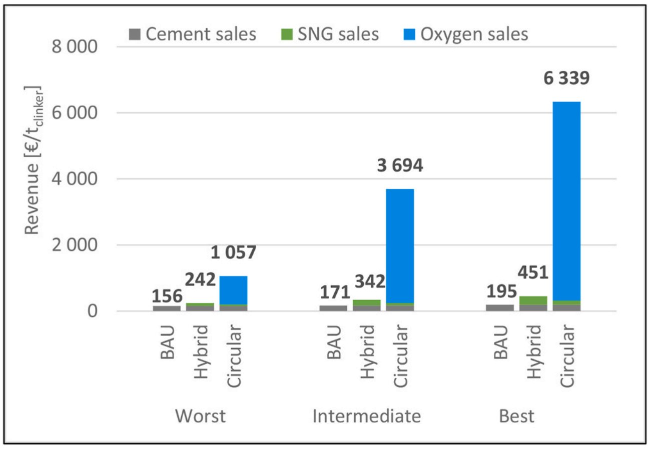

Fig. 5. Revenue per clinker tonne [€/$t_{clinker}$].

sum of cement, oxygen and SNG sales, based on their respective prices, in Table 2, and the quantity produced.

The economic KPIs are summarized in Table 5.

The NPV, in €/$t_{clinker}$, is highly sensitive to the model and scenario. In the intermediate and best-case scenarios, the NPV of both the BAU and circular models is positive, indicating their economic viability by generating a financial surplus in addition to recovering the investment (BAU CAPEX) and fulfilling the minimum income required by investors. Nevertheless, in the worst-case scenario, the NPV of all the models is negative, with the circular model having the lowest value, followed by the hybrid and then the BAU models. Contrastingly, in the other scenarios, the circular model presents the highest NPV, benefiting from a significant increase in revenue, largely driven by the higher O_2 prices, and reduced total costs.

The hybrid model did not generate a cash flow greater than the initial investment in the worst and intermediate scenarios, resulting in an indeterminable IRR. The NPV and IRR results were consistent as a negative NPV corresponds to an IRR lower than the discount rate, and vice versa. The BAU model had the highest IRR in the best-case scenario, while the circular model had the highest value in the intermediate scenario.

Payback periods were compared for each scenario. However, in some scenarios, the payback period of the hybrid and circular models exceeds the project's total lifetime and is therefore not displayed. The payback period values are generally consistent with the obtained IRR results, with a higher IRR corresponding to a lower payback period and vice versa. In the worst-case scenario, only the investment in the BAU model can be recovered within the project's lifetime. In the intermediate scenario, the BAU and circular models have a similar payback period of around 6 years. However, in the best-case scenario, the BAU model has the shortest payback period of 2.1 years, while the circular model takes 3.5 years, and the hybrid model exhibits the longest payback period of 9.6 years, with the latter being the only scenario where the payback period falls below the project's total lifetime.

5.2. Environmental KPIs

Fig. 6 shows the two methods used to calculate the "Total Net CO_2 Emissions". The "Literature" method shows slightly lower calcination CO_2 emissions (0.51 $t_{CO2}/t_{clinker}$) compared to the "Real/Adjusted" method (0.52 $t_{CO2}/t_{clinker}$), due to the higher CaO content in the Portuguese cement industry than that assumed in the IPCC guidelines. Conversely, the "Literature" method's fuel combustion CO_2 emissions are higher than in the "Real/Adjusted" method, which can be explained by the higher fuels' average emission factor assumed.

The "Total Net CO_2 Emissions" and "Total Net CO_2 Avoided" KPIs calculated for each model and scenario are summarized in Fig. 7.

The hybrid model produced a "Total Net CO_2 Emissions" of 0.07–0.08 $t_{CO2}/t_{clinker}$, while the circular model achieved near net-zero

Table 5
Economic KPIs. ND=Not determined.

Scenario	Worst			Intermediate			Best		
System/Model	BAU	Hybrid	Circular	BAU	Hybrid	Circular	BAU	Hybrid	Circular
NPV [€/$t_{clinker}$]	−67	−7042	−7369	145	−2805	14 171	434	217	34 462
IRR [%]	5%	ND	−1%	16%	ND	22%	49%	11%	43%
Payback Period [years]	14.5	ND	ND	6.2	ND	5.5	2.1	9.6	3.5

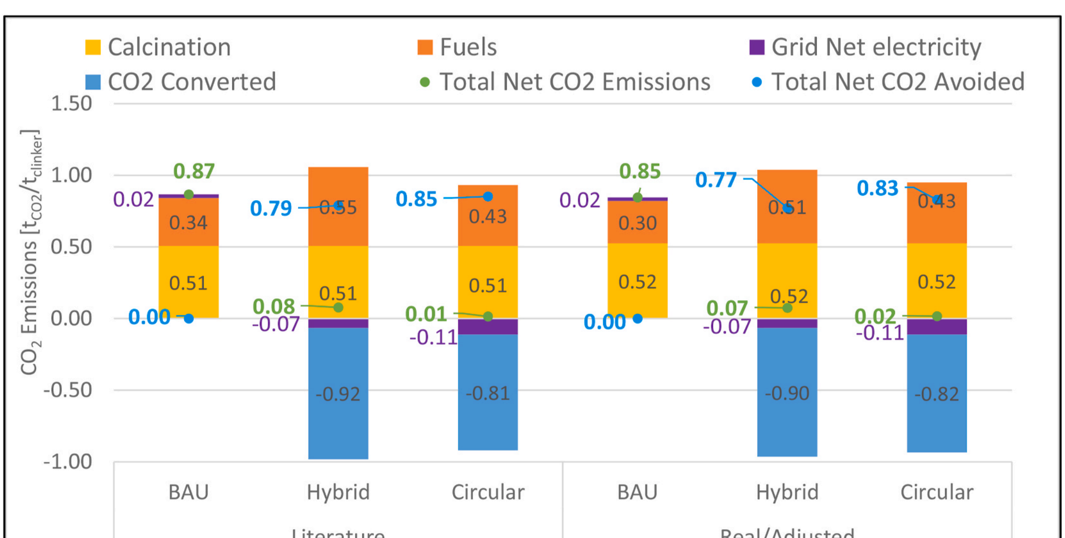

Fig. 6. CO_2 emissions per clinker tonne [$t_{CO2}/t_{clinker}$].

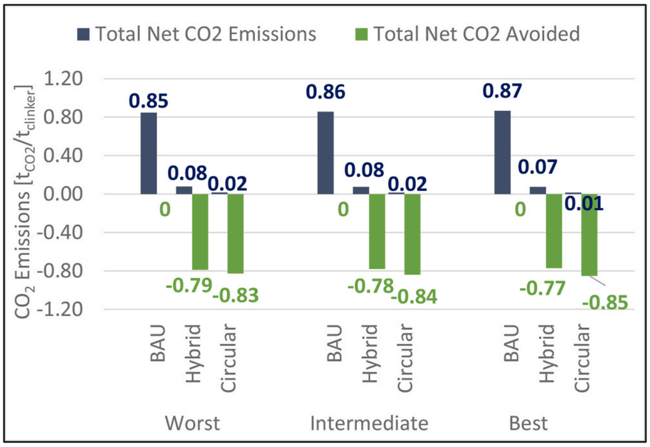

Fig. 7. "Total net CO_2 emissions" and "total net CO_2 avoided" KPIs [$t_{CO2}/t_{clinker}$].

emissions of 0.01–0.02 $t_{CO2}/t_{clinker}$. Although both models exhibit increased fuel combustion CO_2 emissions compared to the BAU model, they convert CO_2 into SNG and produce more non-renewable electricity in the CaL unit than they consume, resulting in negative CO_2 emissions. The circular model captures and reuses CO_2 emissions after conversion into SNG within the cement plant, leading to reduced fuel combustion CO_2 emissions compared to the hybrid model. Furthermore, the circular model required less non-renewable electricity, as it does not include an ASU to produce the O_2 required for the CaL unit. Consequently, the circular model achieved a higher "Net CO_2 avoided" of 98% (0.83–0.85 $t_{CO2}/t_{clinker}$) compared to the 91% in the hybrid model (0.77–0.79 $t_{CO2}/t_{clinker}$). These values could be translated into an annual total net CO_2 avoided emissions up to 0.86 Mt_{CO2} (hybrid model) and 0.92 Mt_{CO2} (circular model). This reduction is significant considering the estimated total annual CO_2 emissions of the three largest Portuguese cement factories (0.92–0.94 Mt_{CO2}, including direct and indirect emissions). The circular mode presents therefore a great potential as a sustainable solution for the cement industry.

Nevertheless, analysing each model's "Net SPEC" as an individual environmental KPI is crucial, considering the significant increase in energy consumption associated with implementing the circular and hybrid models in a cement plant. The hybrid model's heat requirements in the methanation reaction would cause a sevenfold increase in energy consumption, while the circular model's consumption would increase by around fifteen times the BAU value, as shown in Fig. 8. The difference between these two models is attributed to the added electricity

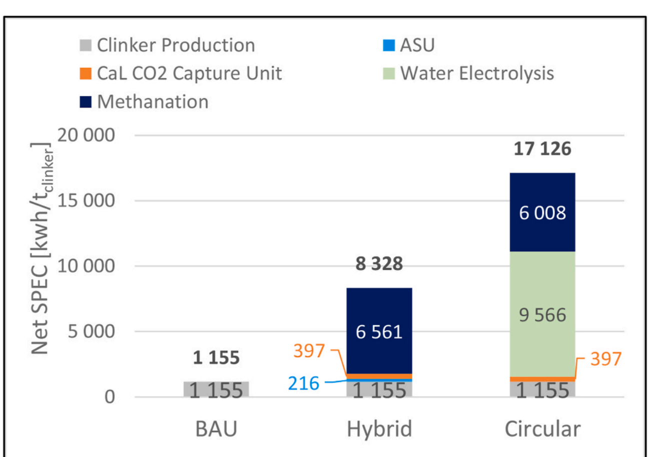

Fig. 8. "Net SPEC" KPI [$kWh/t_{clinker}$].

consumption required to split water into H_2 and O_2, which is not factored into the hybrid model as it occurs beyond the defined boundaries. Despite electricity generation in the "CaL CO_2 capture" covering the demand of the CO_2 capture process and clinker production, these models' energy consumption increases pose a significant challenge.

5.3. Technical KPIs

The technical evaluation is based on three KPIs. The "Direct CO_2 Converted" is 0% in the BAU, as expected considering the KPI's definition, whereas the hybrid and circular models present similar values, around 86%, regardless of the scenario. Table 6 identifies the TRL of the main systems. The technological maturity KPI of each model is determined by the lowest TRL of its constituent process units, which is 9 for the BAU and 6 for both hybrid and circular models.

The "Energy Independence" KPI is 0%, 29% and 66% for the BAU, circular and hybrid models, respectively. The circular model's lower KPI is attributed to its higher "Net SPEC", despite an increase in the on-site solar PV electricity production.

5.4. Eco-efficiency KPIs

The eco-efficiency KPIs and auxiliary calculations are summarized in Table 7.

The total added costs of the hybrid and circular models compared to the BAU were estimated to calculate the "Net CO_2 Avoided Cost" eco-efficiency KPI. Although the circular model achieved a superior "Total Net CO_2 Avoided" environmental KPI compared to the hybrid model, the "Net CO_2 Avoided Cost" KPI was higher due to its increased costs. The CO_2 price, assumed as the BAU's "Net CO_2 Avoided Cost", is significantly lower than the alternative models. However, this KPI does not account for the additional revenue generated by the hybrid and circular models and is therefore insufficient to assess the models' eco-efficiency.

To fill this gap, the "Net Cost of CO_2 Abatement" was calculated, accounting for the net added costs. The importance of the added revenue in the analysis is demonstrated by the discrepancy between the total net added costs and the total added costs, especially in the circular model, where O_2 sales supplement SNG's. The extra revenue is sufficient to cover the added costs for the hybrid model (in the best-case scenario) and for the circular (in the intermediate and best-case scenarios), represented by the negative total net added costs. This difference is translated into the second eco-efficiency KPI, which clearly shows that the implementation of the circular model could result in profit per CO_2 avoided, instead of a cost. Nevertheless, both eco-efficiency KPIs are highly sensitive to scenario changes.

5.5. Multi-criteria decision-making

The AHP method was used to interpret the KPI results for each model and scenario. The indicators were weighted according to their importance, considering the inputs from Portuguese cement companies' stakeholders with a long-term vision as shown in Fig. 9. Evaluation of the consistency ratio (CR) was always below 0.1, indicating the consistency and validity of the AHP analysis [83,84]. Each KPI was normalized depending on the objective (maximize or minimize), as

Table 6
TRL of the different Systems. NA=Not applicable.

System	BAU	Hybrid	Circular	Reference
Clinker Production	9	9	9	The highest TRL (9) was assumed
CaL CO_2 Capture	NA	7	7	[79]
AEL Water Electrolysis	NA	NA	8	[80,81]
Methanation	NA	6	6	[10,82]
Minimum TRL	9	6	6	NA

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Table 7
Eco-efficiency KPIs.

		Worst			Intermediate			Best		
		BAU	Hybrid	Circular	BAU	Hybrid	Circular	BAU	Hybrid	Circular
Auxiliary Calculations [€/$t_{clinker}$]	Total Added Costs	0	676	923	0	388	613	0	194	372
	Total Net Added Costs	0	590	22	0	216	−2909	0	−63	−5773
Eco-efficiency KPIs [€/t_{CO2}]	Net Cost of CO_2 Abatement	59	747	26	90	277	−3459	121	−81	−6770
	Net CO_2 Avoided Cost	59	855	1112	90	496	729	121	251	436

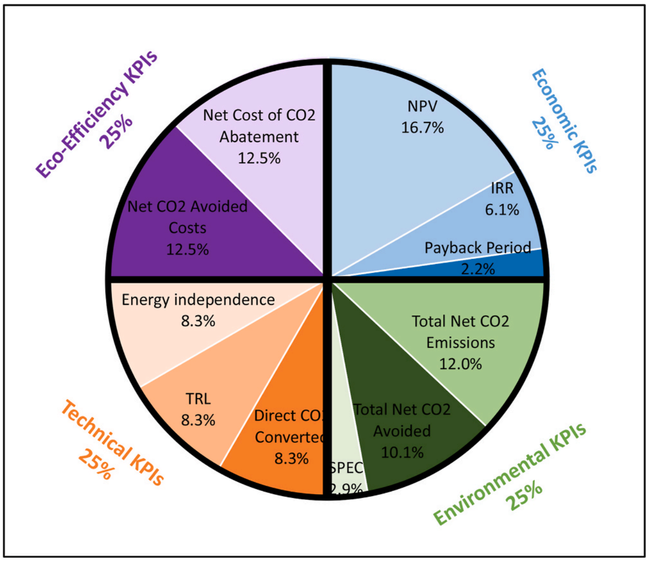

Fig. 9. wt of each KPI.

outlined in the Supplementary Information.

Individual KPIs weighted values were divided into the four main categories. The ranking of the options, from the highest (1st place) to the lowest (3rd place), was determined by the sum of the weighted score of all categories (see Fig. 10). Note that the maximum individual score per category is 0.25.

The hybrid model presented the lowest economic score in all scenarios, with a negative result in the worst and intermediate scenarios, indicating its economic infeasibility compared to the alternatives. This finding aligns with previous studies that deemed a cement CO_2-to-SNG large scale model without SNG internal reuse as "costly and infeasible" [28]. In the worst-case scenario, the BAU model achieved the highest economic score (0.25), whereas the circular model had the highest economic score in the remaining two scenarios. This outcome indicates a promising economic advantage of the circular model over the BAU in most scenarios, while emphasizing the economic competitiveness of a CCU circular cement model compared to a hybrid one.

Environmental KPIs revealed that the circular model outperforms the two alternatives across all scenarios, achieving the highest score of 0.22. This is attributed to the fact that the near net-zero emissions performance offsets its higher "Net SPEC" compared to both alternatives. As expected, the BAU model scored the lowest. Although scenario-specific variations exist in some environmental KPIs, the overall results remain consistent.

The hybrid model's highest technical score (0.22) occurs due to its high energy independence (66%) compared to the circular model (29%) and evidently the BAU (0%). This energy independence reduces the cement plant's relative dependence on the fluctuating energy markets, compensating for its lower TRL (6) compared to the BAU (9). On-site electricity production from the "CaL CO_2 Capture" system and the exported SNG offset the increase in "Net SPEC" compared to the BAU, which is still lower than the circular model. These KPIs are scenario-dependent but yield similar results.

On the contrary, the hybrid model consistently exhibits the lowest eco-efficiency KPIs, across all scenarios. In the worst-case scenario, the BAU attained the highest eco-efficiency (0.17); however, it decreases to 0.12 in the remaining scenarios. The circular model outperforms the BAU with a score of 0.14 and 0.16 for the intermediate and best-case scenarios, respectively. This is a direct consequence of total costs reduction and revenue increase from the worst to the best-case scenarios. While the "Total Net CO_2 Avoided" increases from the worst to the best-case scenario, it has no significant impact on the eco-efficiency KPIs.

In summary, the environmental performance of the circular model remains superior to the alternatives (as expected), with the potential to achieve near net-zero CO_2 emissions, followed by the hybrid and then BAU models. However, the hybrid model scored highest in terms of technical performance, with the circular and BAU models following in

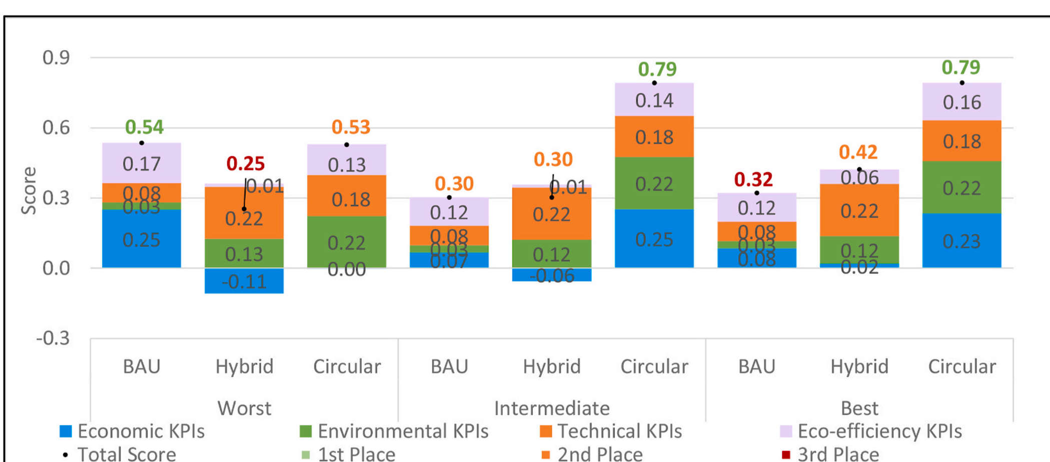

Fig. 10. Score of each model (total and by category).

that order. When considering economic viability, the BAU model showed the highest potential under the worst-case scenario while the circular model outperforms it significantly in all other scenarios. In contrast, the hybrid model constantly ranked as the least economically viable model. These economic findings were supported by the eco-efficiency evaluation, where the BAU model demonstrated the highest eco-efficiency score under the worst-case scenario, while the circular model presented the highest eco-efficiency KPIs score for the remaining scenarios.

Overall, the circular model should be implemented after full validation, within the intermediate or best-case scenario conditions, to achieve the goals defined by the cement industry as it ranks highest in these scenarios. The hybrid model is advantageous over the BAU only within the best-case scenario, and the BAU model could be maintained within the worst-case scenario context. The importance of considering multiple criteria in decision-making processes to achieve sustainable and eco-efficient outcomes is highlighted by these results. This approach is particularly useful in complex systems, such as the one analysed. The implementation of the circular model in the cement industry has the potential to achieve significant reductions in CO_2 emissions, ultimately leading to near net-zero emissions, while increasing the industry's energy independence in a cost-effective manner.

6. Conclusion

The cement's production current linear model is a significant CO_2 emitter dependent on volatile energy markets. To address these issues, a novel circular cement model is proposed, integrating a realistic BAU cement model based on primary data, with three P2G system components: a CaL unit to capture cement-based CO_2 emissions; water electrolysis for on-site hydrogen and oxygen production; a methanation unit to produce large-scale SNG using green H_2 and CO_2. SNG fuels the cement kiln and CaL unit in a loop, while O_2 feeds this unit, enhancing the model's circularity. Although these components have been studied individually, this was the first attempt to integrate them with a cement plant with SNG and oxygen reuse, on an industrial scale.

The model was compared against a hybrid alternative with no SNG reuse or on-site H_2 production, both built on top of the BAU model based on data from Portuguese cement plants to obtain a robust and comparative techno-economic and environmental analysis, across three feasibility scenarios. A cradle-to-gate boundary was assumed and 11 KPIs were calculated, divided into 4 categories. The ranking of the models obtained through the WSM was reported for each scenario considering the KPIs' attributed weights. These weights were based on the AHP method considering the goals and preferences of the Portuguese cement companies.

The multi-criteria decision-making process analysis showed that the circular model outperformed the others significantly in all but the worst-case scenario. The most economical and eco-efficient model is either the BAU (worst-case scenario) or the circular model (intermediate and best-case scenarios) while the hybrid model was constantly found to be the least economically viable model. However, it displayed the highest energy independence. The BAU model could be maintained within the worst-case scenario and the hybrid model was only considered advantageous over the BAU if applied within best-case scenario conditions. The proposed circular model aligns with stakeholders' objectives of reducing dependence on volatile energy markets, achieving near net-zero CO_2 emissions, and promoting a profitable circular economy. This model therefore combines economic and environmental performance with energy independence, providing a promising solution for cement industries seeking to reduce their carbon footprint.

This research highlights the usefulness of the proposed methodology in assisting cement companies to develop their investment strategies towards environmentally sustainable practices. It emphasizes the adaptability of the approach to cater to different stakeholder preferences by allowing customizable KPI weights, providing a valuable resource for policy-making. This research applies the integrated TEA and LCA to highlight economic and environmental advantages of the circular cement production model. The significant reduction in CO_2 emissions and enhanced energy independence position the circular model as a viable strategy for the industry, with potential scalability beyond the Portuguese context. The transition to circular cement production, presents a transformative opportunity. Our research provides a critical roadmap for full-scale deployment and industry-wide impact.

Further research should expand on this work to explore the model's broader environmental impacts and the potential synergies between the gas and cement industries in defossilisation. This is a critical area for future studies. Additionally, the techno-economic and environmental viability of a pilot circular cement plant should be assessed, incorporating a comparison of different electrolyser technologies. Furthermore, mass and energy balances ought to be refined using a single software (e.g., Aspen Plus) that analyses the integrated model. Incorporation of workers' supervisors, laboratory work and patents costs as well as O_2 logistics, distribution, and transportation expenses would provide a comprehensive understanding of the circular cement production process. A comparison between the use of excess O_2 assumed to be sold in the current model and its further use in the clinker burning process to reduce fuel consumption should be performed. Finally, the incorporation of an integrated CaL configuration combining CO_2 capture calciner with the calciner in the cement kiln can also be studied and compared to the proposed circular model.

Author contributions

M. Bacatelo: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft; Writing – review and editing.
F. Capucha: Investigation.
P. Ferrão: Writing – review and editing, Supervision.
F. Margarido: Writing – review and editing, Supervision.
J. Bordado: Writing – review and editing, Supervision.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ijhydene.2024.03.028.

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