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Decarbonization of cement production by electrification

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ABSTRACT

This study presents the techno-economic assessment of four electrified cement plants: i) using direct electrification and plasma technologies (eC-pK case); ii) consisting of indirect electrification via H₂ combustion and oxy-combustion of alternative fuels (OC-HK); iii) combining direct electrification, alternative fuels combustion and post-combustion CO₂ capture (eC-afK); iv) consisting in the electrification of the hydraulic Calcium Hydro Silicate production process (e-hCHS). Process modeling in Aspen Plus is used to estimate mass and energy balances and calculate techno-economic key performance indicators. The study finds that all the electrified alternatives achieve high levels of equivalent CO₂ emissions avoidance (87.2%–101.8%), with a trade-off between the electricity demand (604–1341 kWh/t_{clk}) and the amount of captured CO₂ to be handled by the transport & storage infrastructure (357–834 kg_{CO2}/t_{clk}). With an electricity price of 50 €/MWh, the partially electrified alternatives (OC-HK, eC-afK) showed competitive additional cost of clinker (87 €/t_{clk}) and cost of avoided CO₂ (101 €/t_{CO2}) against a benchmark case, though higher than the cost of the best CO₂ capture technologies from the literature. The eC-pK case resulted in lower economic performance associated mainly to the higher price of electricity per unit of final energy supplied compared to alternative fuels.

1. Introduction

The production of cement is one of the largest CO2-emitting industries, accounting for approximately 8% of global CO2 emissions (Andrew, 2017). Because the majority of these emissions arise from the decarbonation reaction of CaCO₃ and not from fuel combustion (i.e., process-related emissions), there is consensus that carbon capture and storage (CCS) technologies will play the main role in abating CO2 emissions from cement production, as stated by the European Cement Association (CEMBUREAU, 2020) and reports conducted by the International Energy Agency (IEA, 2019, 2020). Post-combustion capture (PCC) systems using absorption with amines is the most developed CCS technology today. In fact, the Norwegian Government has recently approved the implementation of an amine-based capture system to be implemented in the first full-scale CO₂ capture project in a cement manufacturing plant at the Brevik cement plant as part of the Longship CCS project (Global Cement, 2020). On the other hand, oxyfuel technology has gained momentum after several technical reports highlighted the potential for high capture rates and lowest cost of avoided CO₂ (ECRA, 2012; IEAGHG, 2013). The results led to the advancement of numerous research projects, such as AC²OCEM, with the goal of developing first and second generation oxyfuel technology (AC2OCEM,

2019), or the catch4climate project for building a first generation oxyfuel demonstration plant (CI4C, 2020). Progress has continued and the technology will soon reach full-scale implementation with the operation of two separate projects awarded with the EU Innovation fund, one in France (EQIOM, 2022) and the second one in Germany (Holcim, 2022).

Notwithstanding the fact that CO_2 capture will be necessary to avoid process emissions from $CaCO_3$ calcination, electrification of heat supply is an interesting option to reduce the CO_2 generated from fuel combustion, in locations with access to low-cost and low-carbon electricity. A recent technical report on the decarbonization of cement industry, commissioned by the European Commission, highlighted the benefit of complementing carbon capture technologies with the lower volumes and better quality of emissions resulting from an electrified process (Marmier, 2023).

A report conducted by Cementa and Vattenfall (Cementa and Vattenfall, 2018) found that an electrified cement plant could be economically competitive against a reference cement plant with an amine-based PCC system. The results were based on a mixture of resistive elements and the use of plasma generators to deliver the high temperature heat. To estimate mass and energy balances, the authors resorted to theoretical calculations based on simplified assumptions and experienced-based temperatures at various steps of the process. Likewise, the Mineral

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Product Association (MPA) in collaboration with Cinar and VDZ studied the impact of switching to a mixture of biomass, plasma burners, and indirect electrification through H_2 (MPA, 2019). Using Mineral Interactive Computational Fluid Dynamics (MI-CFD) modelling, the report concluded that a net zero fuel mix is possible but further work is needed on testing the technologies as well as a more comprehensive techno-economic assessment of potential scale up.

Other projects have also been identified in the subject of electrifying the cement manufacturing process. The first one is the Decarbonate project, in Finland, which analyzed the calcination of limestone at an electrically heated rotary kiln (Tsupari et al., 2022). Within the activities, a mobile pilot plant on an electrified rotary calciner with a capacity of >100 kg/h was built, which obtained good results in terms of quality of the calcined material and the concentration of the produced CO₂ (98 %vol.dry). Another project is the Norwegian-based "ELSE", currently in its phase 2 (ELSE - 2), which also focuses on the electrification of the calcination step (CLIMIT, 2020). During this stage, the goal is to design a pilot plant that can later be tested in phase 3. In this framework, partners of the project have recently published a modelling study of a cement plant using an electrified calciner (Jacob and Tokheim, 2023). The focus of the study is on the energy demand and CO₂ emissions, comparing the results under different calciner designs: entrainment calciner (high CO₂ recycle), fluidized bed calciner (low CO2 recycle), and a rotary calciner (no CO₂ recycle). Simulations in Aspen Plus showed that the electrified designs achieve 78% capture rate if all CO₂ emissions from the calciner are captured, with an energy demand increase of 23%, 6.5%, and 1.2% for the high CO2 recycle, low CO2 recycle, and no CO2 recycle, respectively.

The goal of this study is to assess different options of electrification of cement production from a techno-economic perspective through a process engineering study, identifying advantages, disadvantages and the trade-offs of the different technologies. With respect to existing studies in the literature, this work compares for the first time five different electrified processes for cement production and performs a consistent economic comparison of the different electrified processes with benchmark plants with CO_2 capture.

The paper is structured as follows: In Section 2, the methods used to conduct the techno-economic assessment are detailed. First, Section 2.1 presents the reference cement plant and the assumptions for the Aspen model. Then, in Section 2.2, the design of the electrified alternatives, including the configuration of the processes and the main assumptions are provided. The technical and economic results obtained from the process models are discussed in Section 3.1 and Section 3.2, including two sensitivity analysis on the cost of electricity and the cost of CO_2 emissions. Then, Section 3.3 elaborates on the role of electrification and carbon capture. Finally, the conclusions of the study are presented in Section 4.

2. Methods

The methodology to conduct the techno-economic assessment of the electrified cement plants is carried out in three steps. First, the process model of a reference cement plant is built in Aspen Plus v10 using the values from the Best Available Technique (BAT) plant to validate the results (Schorcht et al., 2013). The second step is the design of the electrified cement plants based on different technologies to supply the heat requirements in the pre-calciner and the rotary kiln. This include adapting the process to new configurations and changing the operating conditions in some unit operations. The designs are then translated into the simulation environment to obtain mass & energy balances. Finally, key performance indicators (KPIs) are estimated and the technologies are compared from a techno-economic perspective.

2.1. Reference plant

A steady-state model of the reference cement plant is developed in

Aspen Plus, following the Best Available Technique (BAT) standard defined in the European BREF-Document (Schorcht et al., 2013) and detailed in the CEMCAP project (Voldsund et al., 2019).

Fig. 1 shows a diagram of the plant. The technology is based on a drykiln process, comprising a 5-stage preheating tower, a pre-calciner with tertiary air duct, a rotary kiln, and a grate cooler, with coal as fuel and a capacity of about 3000 ton of clinker per day. The key assumptions used for the modelling of the reference cement plant are provided in the supplementary material.

In addition, a reference cement plant coupled with an absorptionbased PCC system is used as the benchmark decarbonization strategy. MEA is chosen as the solvent for the PCC process, with heat supplied by an air-sourced heat pump with a COP of 2.

2.2. Electrified cement plants

The design of the electrified cement plants is conducted considering information from previous reports (Cementa and Vattenfall, 2018; MPA, 2019) and analyzing the suitability of a range of electrification technologies. From this analysis, two partially electrified and two fully electrified cases are designed:

- i) eC-pK (fully electrified): the first case involves an electrified precalciner using a resistive element or magnetic induction, and plasma gas to deliver the high-temperature heat in the rotary kiln;
- ii) OC-HK (partially electrified): the second case combines oxycombustion of alternative fuels in the pre-calciner coupled with the indirect electrification of the rotary kiln by burning H₂ produced from steam electrolysis;
- iii) eC-afK (partially electrified): the third case combines an electrified pre-calciner with combustion of alternative fuels in the rotary kiln and a solvent-based PCC system;
- iv) e-hCHS (fully electrified): the fourth case is the electrification of the hydraulic Calcium Hydro Silicate (hCHS) production process, based on the Celitement© technology.

A brief description of the electrified cases is provided in the following paragraphs. More information on the modelling methods and the details of the main streams can be found in the supplementary material.

The eC-pK case (Fig. 2) couples resistive or inductive electricity with the use of plasma technology. The basis for the design is drawn from the experience of the CemZero report (Cementa and Vattenfall, 2018). In this case, the heat demand in the pre-calciner is met using a resistive



Fig. 1. Schematic of the reference cement plant.



Fig. 2. Schematic of the eC-pK plant: direct electrification of the calciner coupled with plasma torch in the rotary kiln.

element or through magnetic induction. The same CaCO₃ calcination degree as in the reference plant is considered, heating the raw meal to a higher temperature of 920 °C to account for the higher CO₂ partial pressure. The rest of the calcination and the formation of the clinker phases continues in the rotary kiln, where air is used as plasma gas to deliver the high temperature needs. Plasma burners are used to pre-heat ambient air to 3,470 °C before entering the kiln. The temperature is chosen to match the operating condition of the plasma generators in the CemZero study. The flow of plasma gas, which is mixed with secondary air from the clinker cooler, is controlled to achieve an outlet temperature of the clinker phases of 1,450 °C.

The second alternative (OC-HK) is based on the indirect use of electricity in the rotary kiln (i.e., through the production of green H_2) and oxy-combustion of alternative fuels for the heat demand in the precalciner (Fig. 3). H_2 production through a Solid Oxide Electrolysis (SOEC) system is considered because of its higher efficiency (80–85% electricity to LHV efficiency) and the fact that part of the waste heat in



Fig. 3. Schematic of the OC-HK plant: indirect electrification through $\rm H_2$ combustion in the rotary kiln and oxyfuel combustion of alternative fuels in the calciner.

the process can be utilized to produce the steam requirement. Previous research conducted by the Mineral Product Association has developed this concept. First, in a modelling study that combined plasma technology in the pre-calciner and a mixture of 50% biomass and 50% hydrogen in the rotary kiln (MPA, 2019), followed by a successful trial that demonstrated the feasibility of combusting 39% hydrogen with a mixture of alternative fuels (MPA, 2021). The application of 100% H₂ combustion would entail technical (and possibly safety) issues related to the different combustion properties, such as wide flammability limits, burning velocity and short flame(Cementa and Vattenfall, 2018), which might require some modifications on conventional burners (Sandalow et al., 2019). However, for the purpose of this study, it is assumed that the technological and operational problems of burning pure H₂ in the rotary kiln can be solved.

The waste heat available is recovered to supply steam to the SOEC and use the surplus for any other heat demand. The SOEC is assumed to operate at 700 °C (Hauch et al., 2020) and to produce H₂ with a specific electricity consumption of 40 kWh/kg_{H2} (i.e. 83.3% electricity to LHV conversion efficiency) (Nechache and Hody, 2021). A share of the H₂ is recycled back and mixed with the inlet steam to avoid oxidation of the cathode materials. This recycle is controlled to reach an inlet concentration of 10/90 %mol of H₂/H₂O at the cathode inlet (Bianchi and Bosio, 2021; Kim et al., 2016). A steam utilization factor of 90% is assumed considering the higher end of the operating range found in the literature (Bianchi and Bosio, 2021; Posdziech et al., 2019).

The third alternative (eC-afK) consists of an electrified calciner, a rotary kiln burning alternative fuels, and a PCC system using monoethanolamine (MEA) to capture the CO₂ emissions from the kiln (Fig. 4). Similar to the first case, electric power is supplied to the calciner to achieve a temperature of 920 °C by either a resistive element or magnetic induction. On the other hand, the heat demand for the rotary kiln is delivered by burning alternative fuels. The solvent-based PCC unit is designed to capture 95% of the CO₂ emissions from the rotary kiln, using the available waste heat from the new configuration to supply heat to the regeneration unit.

The last alternative (e-hCHS) is based on the production of hCHS (Fig. 5), a novel approach producing a cementitious binder with similar mixing, setting, and hardening characteristics as standard Ordinary Portland Cement (OPC) (Stemmermann et al., 2010). The electrification of the reference process assumes some modifications, particularly in the drying stage (Fig. 6). The equipment is operated with superheated steam



Fig. 4. Schematic of the eC-afK plant: electrified calciner and combustion of alternative fuel in the rotary kiln, with MEA-based CO₂ PCC.



Fig. 5. Schematic of the reference hCHS production process.



Fig. 6. Schematic of the electrified hCHS production plant.

instead of air, similarly to WTA lignite drying process (Klutz et al., 2011). In this case, part of the steam (stream 6 in Fig. 6) is compressed to 4 bar and used as drying agent by condensing in tubes through a steam heat pump system, while the rest is used as fluidization medium. In such drying process, the energy required to remove the moisture can be supplied entirely from the evaporated and circulated moisture of the material, without requiring an additional steam make-up (Kakaras et al., 2002). This reduces the energy consumption only to the compression of the drying and of the fluidization vapor. It has to be noted that the WTA process considers a fluidized bed solid-vapor contactor. In case CSH particles cannot be fluidized, the same principle could be applied in driers with different geometry, such as tube-bundle driers, with minor impact on the energy consumption of the process. For the autoclave step, an electric boiler with 99% efficiency is assumed to deliver the steam requirements.

3. Results and discussion

3.1. Technical results

The summary of the heat and electricity demand of the assessed plants is presented in Table 1. The first section shows the thermal energy demand to sustain the calcination reaction, the formation of the clinker phases, and/or any other specific heat demand in the process. The second section shows the electricity required to supply the heat in the electrified alternatives, with different efficiencies depending on the technology, and any auxiliary energy needs. It is important to differentiate between direct variations in fuel consumption and the heat demand. For instance, while the reference case with MEA PCC system has the largest increase in thermal energy needs (+95.8% with respect to the reference case), the direct fuel consumption remains the same as in the reference cement plant. This is because the additional energy required to regenerate the solvent is assumed to be supplied by a heat pump.

The largest reduction in direct fuel consumption is given by the fully electrified alternatives, i.e., cases eC-pK and e-hCHS (-100%), followed by the partly electrified cases, eC-afK (-57.4%) and OC-HK (-7.3%).

Likewise, the electrified alternatives eC-pK, OC-HK, and eC-afK result in an increase of total heat demand (+18.1–23.9%). This is related to a decay of the overall thermal efficiency of the electrified processes, caused by different factors: (i) tertiary air no longer complements the heat demand in the pre-calciner, (ii) additional heat is required in the OC-HK case for heating the CO₂ recycle stream up to the calcination temperature, (iii) in the OC-HK and eC-afK cases, a lower LHV fuel is used in the calciner and the rotary kiln, respectively and (iv) the plasma generator of the eC-pK case involves higher primary/secondary air ratio (i.e. less efficient use of waste heat) in the rotary kiln.

It must be observed that in the proposed configurations, waste heat from the hot CO_2 -rich stream and the mixture of unused tertiary air and vent air is available. In the eC-afK case, this waste heat is harnessed internally for the production of steam to reduce the energy requirement from the heat pump. In the other cases, the amount of waste heat available is presented in Table 1, but no valorization strategies have been included for the techno-economic evaluation. The available waste heat is estimated up to a temperature of 135 °C. If we consider a recovery cycle with a heat-to-electricity conversion efficiency of 30%, it would be enough to supply 2.8% and 8.1% of the total electricity demand of the eC-pK and OC-HK cases, respectively.

The increase in the electricity demand in each case depends both on the electrified heat duty and on the efficiency of the technologies used to deliver the heat requirements (95% for direct electrification, 85% for plasma generator, 83% for SOEC, and 200% for heat pump; see supplementary material). Taking everything into consideration, the electrification of the cement manufacturing process supported by the different technologies in the eC-pK, OC-HK, and eC-afK cases translates into an electricity demand increase of 10.2, 4.5, and 7.1 times the reference (i.e. a net demand of 1,341, 598 and 929 kWh_e/t_{clk}), respectively.

In the e-hCHS case, the optimization of the drying stage results in a 12% reduction of the heat demand in hCHS production when compared to the reference process. Under the new configuration, 50% of the evaporated and recirculated steam is used as fluidization medium and 50% as drying agent. The outcome of the simulation shows that this is enough to supply 94.5% of the drying heat needed. The rest of the drying steam is supplied with an electric boiler (stream 8 in Fig. 6).

The total electricity consumption in the e-hCHS case is completed with the demand from the autoclave, the grinding of sand, the

Table 1

Summary of heat and electricity demand.

Energy		Reference case	Reference case with PCC	eC-pK	OC-HK	eC-afK	hCHS-ref	e-hCHS
Heat								
Heat demand in calciner	GJ _{th} /t _{clk}	1.99 ^F	1.99 ^F	2.38^{E}	2.97^{AF}	2.38^{E}	_	_
Heat demand in rotary kiln	GJ _{th} /t _{clk}	1.21^{F}	$1.21^{\rm F}$	1.41^{E}	1.00^{H}	1.37 ^{AF}	_	_
Heat demand for MEA regeneration	GJ _{th} /t _{clk}	-	3.07 ^{HP}	-	-	0.16^{HP}	_	_
Heat demand in PFRK (lime)	GJ _{th} /t _{clk}	-	_	-	-	-	1.64 ^F	1.64 ^E
Heat demand hCHS process	GJ _{th} /t _{clk}	-	_	-	-	-	1.40 ^F	1.23^{E}
Net waste heat available ^a	GJ _{th} /t _{clk}	-	_	0.45	0.58	0.49^{b}	_	_
Total heat demand	GJ _{th} /t _{clk}	3.21	6.27	3.78	3.97	3.90	3.03	2.87
Change w/r to reference	%	-	+95.8%	+18.1%	+23.9%	+21.8%	-5.4%	-10.5%
Total direct fuel consumption	GJ _{th} /t _{clk}	3.21	3.21	0	2.97	1.37	3.03	0
Change w/r to reference	%	-	0%	-100%	-7.3%	-57.4%	-5.4%	-100%
Electricity								
Electrified calciner	GJ _{el} /t _{clk}	-	_	2.50	-	2.51	-	_
Plasma burners	GJ _{el} /t _{clk}	-	_	1.65	-	-	-	_
Heat pump MEA system	GJ _{el} /t _{clk}	-	1.53 ^c	-	-	0.08 ^c	-	_
Air Separation Unit (ASU)	GJ _{el} /t _{clk}	-	_	-	0.11	-	-	_
CO2 compression and purification	GJ _{el} /t _{clk}	-	0.27	0.20	0.37	0.26 ^d	-	0.13
SOEC	GJ _{el} /t _{clk}	-	_	-	1.20	-	-	_
Electrified PFRK - Lime production	GJ _{el} /t _{clk}	-	_	-	-	-	-	1.72
hCHS - Tribochemistry step	GJ _{el} /t _{clk}	-	_	-	-	-	1.44	1.44
hCHS - Electrified heat supply	GJ _{el} /t _{clk}	-	_	-	-	-	-	0.92
Other auxiliaries	GJ _{el} /t _{clk}	0.47	0.62	0.47	0.47	0.51	0.27	0.27
Total electricity	GJ _{el} /t _{clk}	0.47	2.43	4.83	2.15	3.35	1.71	4.48
	kWh _{el} /t _{clk}	131.7	675.5	1,341.0	597.6	929.1	474.6	1,245.5
Final energy								
Total energy demand (fuel + electricity)	GJ/t _{clk}	3.7	5.6	4.8	5.1	4.7	4.7	4.5

Heat is supplied from different sources: "F" = fossil fuels; "HP" = heat pump; "E" directly electrified or e-boiler; "AF" = alternative fuels; "H" = H_2 from electrolysis. The heat demand from fossil fuels, alternative fuels, and H_2 is based on LHV.

^a Heat available from the hot CO₂ stream and from the mixture of III and vent air at temperatures above 135 °C.

^b Waste heat in the eC-afK case is used internally to reduce the heat requirement from the heat pump.

^c Calculated considering a COP of 2.

^d Includes electricity consumption from the CO₂ Compression and Purification Unit (CPU) and the CO₂ compression in the MEA PCC system.

mechanochemical activation, and others. The first thing to notice is that the heat demand is 5.4% and 10.5% less in the hCHS and e-hCHS production processes, respectively, because of the reduced CaO requirement and the efficiency improvement in the drying step of the electrified alternative. This reduction in heat demand is contrasted with a 3.6x and 9.5x increase in the electricity consumption for the hCHS-ref and ehCHS cases, respectively, compared to the reference cement plant.

The trade-off between direct fuel consumption and electricity demand is more clearly represented in Fig. 7. The reference process lays in the bottom right corner, with the lowest electricity demand. As the heat supply is electrified, the electricity demand increases while the fuel consumption decreases. From the figure, the alternatives can be clustered in three different groups based on their impact in the fuel consumption. In the first cluster, the Ref + MEA, hCHS-ref, and OC-HK cases show only a slight or no reduction in the fuel consumption, even though the electricity demand is increased in the range of 3.6x - 5.1x. In the second cluster, the eC-afK case already shows a significant reduction of



Fig. 7. Trade-off between total fuel consumption and total electricity consumption.

57% in fuel requirement with respect to the reference, in exchange of a higher electricity demand. Finally, the fully electrified alternatives eCpK and e-hCHS show both the largest decrease in direct fuel consumption (to zero) and the highest increase in electricity demand. Overall, looking at how the low carbon cases are positioned in this chart, a total energy demand (i.e. electricity + fuel) of about 4.5–5.1 GJ/t_{clk} is obtained, meaning an electricity to fuel substitution in the range of 0.8–1.1 GJ_e/GJ_{LHV}.

In electrified processes, CO_2 emissions must be evaluated considering both direct and indirect emissions. Direct CO_2 emissions (Scope 1) correspond to the CO_2 produced during calcination of the raw material, including emissions from the decarbonation of the limestone, and fuel combustion when applicable. Indirect CO_2 emissions (Scope 2) arise from the electricity consumption. In this work, we assume a grid with a carbon intensity of 50 kg_{CO2}/MWh as the baseline (representative of grids dominated by low-carbon power generation technologies). The summary of CO_2 emissions from each technology and their source is presented in Table 2.

The direct CO₂ emissions of the electrified alternatives are influenced by the calcination rate in the pre-calciner, the capture efficiency of the CPU and the MEA system, and the use of carbon neutral fuel. In the eCpK and eC-afK cases, the CPU is assumed capable of capturing 100% of the CO₂ generated in the pre-calciner, considering that the CO₂ is not diluted by flue gas from fuel combustion and that the ingress of false air is considerably reduced. On the other hand, the mass and energy balance of the CPU in the OC-HK case is based on the work by (Magli et al., 2022) for calcination under oxy-combustion conditions. Specifically, the initial CO₂ purity of 89.6%vol. dry, involves the loss of 0.7% of the CO₂ at the CPU inlet, i.e. 5.8 kg_{CO2}/t_{clk} . Moreover, the alternative fuel burned in the OC-HK and eC-afK cases is assumed to be sourced with a biogenic content of 30%, i.e., 30% of the CO₂ emissions from combustion are neutral if emitted or carbon negative if captured and stored. Coupled with the high capture rates of the CPU (99.3%-100%) and the MEA system (95%), the OC-HK and eC-afK alternatives are capable of

Table 2

Summary of direct and indirect CO2 emissions.

CO ₂ emissions		Reference case	Reference case with PCC	eC-pK	OC-HK	eC-afK	hCHS – ref	e-hCHS
Direct CO ₂ emissions								
CO ₂ from calciner (to CPU)	kg _{CO2} /t _{clk}	712.9	712.9	536.4	840.4	534.0	_	_
CO ₂ concentration in calciner string	%vol.dry	-	_	99.0%	89.6%	99.0%	-	_
CO ₂ from lime production	kg _{CO2} /t _{clk}	-	_	-	-	-	498.8	365.5
CO2 emitted from CPU vent	kg _{CO2} /t _{clk}	-	_	-	5.8	-	-	_
CO ₂ from rotary kiln	kg _{CO2} /t _{clk}	146.7	146.7	43.9	39.0	182.6	-	-
CO ₂ concentration from kiln string	%vol.dry	32.7% ^a	32.7% ^a	2.9%	7.2%	15.0%	-	-
CO ₂ from NG combustion – hCHS	kg _{CO2} /t _{clk}	-	-	-	_	-	76.8	-
Biogenic CO ₂ generated	kg _{CO2} /t _{clk}	-	-	-	90.4	41.5	-	-
Captured CO ₂	kg _{CO2} /t _{clk}	-	816.6	536.4	834.6	707.4	_	356.5
Captured CO ₂ to storage	t _{CO2} /year	-	765,034	502,413	781,204	662,589	_	333,862
Direct CO ₂ emissions	kg _{CO2} /t _{clk}	859.6	43.0	43.9	-45.6	-32.4	575.6	-
CCR direct emissions	kg _{CO2} /t _{clk}	-	95.0%	92.4%	94.9%	98.7%	_	100%
Direct CO ₂ emissions avoided	kg _{CO2} /t _{clk}	-	95.0%	94.9%	105.3%	103.8%	_	100%
Indirect CO ₂ emissions								
Indirect CO ₂ emissions	kg _{CO2} /t _{clk}	6.6	33.8	67.1	29.9	46.5	23.7	62.3
Net CO ₂ emissions								
Net equivalent CO ₂ emissions	kg _{CO2} /t _{clk}	866.2	76.8	110.9	-15.7	14.1	599.4	62.3
Equivalent CO ₂ avoided	kg _{CO2} /t _{clk}	-	91.1%	87.2%	101.8%	98.4%	_	92.8%

^a The configuration of the reference cement plant only has 1 preheating string. The CO₂ concentration at the top of the kiln string applies for both the CO₂ emitted in the rotary kiln and in the calciner.

generating negative direct CO_2 emissions. Because of this characteristic, the technologies achieve a ratio of direct emissions avoided superior to 100% (105.3% and 103.8%, respectively). Nonetheless, this indicator has to be interpreted with the amount of captured CO_2 . Indeed, higher flowrates of captured CO_2 involve larger infrastructure and higher absolute cost for CO_2 management. In this sense, the largest flowrate of captured CO_2 is calculated for the OC-HK case, followed by the MEA case. The larger flowrate of CO_2 emissions in the OC-HK compared to the reference case, despite having a lower fuel consumption, is a consequence of the 9% increase in the carbon/LHV ratio of the alternative fuel with respect to coal. Finally, in the e-hCHS alternative, all the direct CO_2 emissions are generated during the lime production, where also 100% capture is assumed due to the extremely high CO_2 concentration expected for the gas from an electrified lime calciner.

When the indirect emissions are included, the eC-pK case exhibits a lower potential than the benchmark in equivalent CO_2 emissions avoided. Nevertheless, it is worth noting that this alternative is coupled with a 34% reduction in captured CO_2 , with the subsequent impact in the handling, transporting, and storage of the CO_2 . Similarly, the e-hCHS case could mean an additional 22% reduction under the assessed conditions, by only capturing the CO_2 emitted during lime production. The eC-afK alternative also reduces the amount of captured CO_2 with respect to the benchmark, but at a lower rate of 13%. On the contrary, the OC-HK case has the highest amount of captured CO_2 , exceeding the estimation for the benchmark by 2%.

Fig. 8 shows the amount of captured CO_2 versus the electricity consumption in each technology. Because all the alternatives have high

capture rates, the figure helps to identify trends regarding the degree of electrification with respect to the CO_2 capture and transport infrastructure needed to achieve high levels of CO_2 emissions avoided. Indeed, the findings suggest a trade-off between the electricity consumption and the amount of captured CO_2 , with the electrified hCHS production process resulting in the lowest flowrate of captured CO_2 , followed by the fully electrified eC-pK case. On the contrary, while the partially electrified OC-HK case and the benchmark both need roughly 50% of the electricity from the fully electrified alternative, they also result in the largest amount of captured CO_2 .

Analyzing the trend between all alternatives except the e-hCHS case, a relationship of $-0.41 \text{ kg}_{\text{CO2}}/\text{kWh}$ is found. This means that for every additional kWh of electricity, there is a reduction of 0.41 kg of captured CO₂ that needs to be managed. Compared to the rest, the e-hCHS case is an outlier. If the trend between the e-hCHS and the Ref + MEA cases is considered, then the absolute value of the ratio increases to 0.8 kg_{CO2}/ kWh, doubling the reduction in captured CO₂ to be handled for every kWh of additional electricity compared to the other alternatives.

The impact of the carbon intensity of the power grid on the CO_2 emissions is assessed through a sensitivity analysis. The results can be seen in Fig. 9. The four electrified technologies are shown in coloured lines, the reference hCHS production is shown in black, the reference cement plant is shown in a straight grey line, and the benchmark using PCC with MEA is depicted in a dotted grey line. Two sections representing the carbon intensity of a low-carbon grid (0–50 kg_{CO2}/MWh)





Fig. 8. Total electricity consumption versus captured CO2-

Fig. 9. CO₂ emissions as a function of the carbon intensity of electricity.

and the carbon intensity of a Natural Gas Combined Cycle generation plant (330-400 kg_{CO2}/MWh) are also included as point of reference. The figure highlights the dependency of the electrified alternatives to the carbon intensity of the grid, with the eC-pK and e-hCHS cases showing the steepest slopes. It also illustrates that all the electrified cases can considerably reduce the carbon footprint of the reference processes, even under a fossil fuel-based energy grid. Nevertheless, the three alternatives compare differently against the benchmark decarbonization technology. The direct capture rate of eC-pK is equal to the MEA case but it needs twice as much electricity, therefore, the carbon footprint is always higher. In the e-hCHS alternative, less CO2 is emitted under a renewable/nuclear scenario, with the crossover point found at a carbon intensity of 75 kg_{CO2}/MWh. Similarly, but through a larger range, the eC-afK case outperforms the benchmark up to a carbon intensity of 297 kg_{CO2}/MWh. Finally, the OC-HK alternative always stays below the benchmark and all the other cases, because of the negative emissions arising from the combustion of alternative fuels with CO₂ capture and because of the comparatively low specific electricity consumption, leading to the mildest slope among the low-emission cases.

3.2. Economic analysis

The economic performance of the electrified alternatives is analyzed by calculating the following KPIs: CAPEX, OPEX, cost of avoided CO_2 (CAC), and additional cost of clinker (COC). Results for the e-hCHS case are not included, given that there is no available information in the open literature to estimate the equipment costs.

The breakdown of the CAPEX for the 4 assessed alternatives is presented in Fig. 10. The methodology used was obtained from the work of (Gardarsdottir et al., 2019) and more details are provided in the supplementary material. The largest increase in equipment cost comes from the Ref + MEA case, highly influenced by the cost of the heat pump system used to produce the heat for the regeneration of the solvent, representing 2/3 of the 277 M€. The second technology with the highest CAPEX is the OC-HK case, driven primarily by the cost of the electrolyzer (42.7%) and the CPU (38.3%), which is 34% more expensive compared with the other alternatives due to the larger size needed to handle the higher flowrate of captured CO₂. The rest is completed with the ASU (18.0%) and a small fraction of other minor investments representing 1% of the overall cost. In the eC-pK case, the CAPEX is comprised of the plasma generator system (40.4%), the CPU (34.5%), and the electrified calciner (25.1%). For the electrified calciner, the cost of a conventional unit is scaled from the work of (De Lena et al., 2019), and an additional 100 €/kW is assumed for its electrification. Finally, the CAPEX of the eC-afK alternative is comprised of the CPU (42.5%), the electrified calciner (31.0%), the equipment from the MEA system (20.7%) and finally, the heat pump (5.8%).

Fig. 11 depicts the breakdown of the additional COC with respect to the reference process without CO_2 capture. The reference case is also



Fig. 10. Breakdown of additional CAPEX with respect to the reference plant without CO_2 capture.



Fig. 11. Breakdown of the additional COC with respect to the reference plant without CO₂ capture.

included as comparison basis, where the incremental costs are associated to the CO₂ emissions, with a carbon tax of 100 ℓ/t_{CO2} . The results highlight the relevance of the CAPEX, the electricity cost (yellow patterns), and the influence of the transport and storage (T&S) of the captured CO₂. An important share of the additional COC is related to the CAPEX. Depending on the technology, the CAPEX and fixed costs (calculated as a function of the CAPEX) represent between 27% and 45% of the additional COC, with the largest share found for the OC-HK case. The technology more reliant on electrification, i.e., the eC-pK case, shows the highest OPEX. The bulk of the OPEX is primarily attributed to the electricity demand from the electrified calciner and the plasma generators, representing 27% and 18% of the total additional COC, respectively. The electrified calciner also plays a major role in the eC-afK case, with a 34% share of the additional COC. On the other hand, the electricity demand from the Ref + MEA and the OC-HK cases is mainly given by the heat pump and the H₂ production, accounting for 20% and 19% of the respective additional COC. Furthermore, the CO₂ tax and the T&S of captured CO₂ play a significant role in each case. Together, these items represent 27%, 22%, 22%, and 22% of the additional COC for the Ref + MEA, eC-pK, OC-HK, and eC-afK cases, respectively, where in the OC-HK case, the CO₂ tax represents a revenue. These underscore the relevance that the capture of negative emissions and the cost of T&S play in the technologies with higher amount of captured CO₂.

It is also important to notice that the use of available waste heat was not included in the economic performance of the eC-pK and OC-HK cases. Indeed, because of the new configurations, the unused heat from the hot CO₂-rich outlet stream and from the III and vent air could be harnessed to operate a recovery cycle to supply a share of the electricity demand. However, the impact would be minimal. As previously discussed, assuming a recovery system with 30% efficiency, the electricity demand could be reduced by 2.8% and 8.1% in the eC-pK and OC-HK cases, respectively. This would translate into a reduction in the additional COC of 1.5% and 2.2%, respectively, without considering the



Fig. 12. Breakdown of the cost of avoided CO2 (CAC).

increment in CAPEX.

Fig. 12 shows the details of the CAC. The breakdown underscores the relevance of the CAPEX and the electricity cost. In particular, the eC-pK case has the highest share of the CAC associated to electricity consumption (58%), where the majority comes from the electrified calciner followed by the plasma generators. In the other cases, the share of the electricity consumption starts to decrease from 47% for the eC-afK case, influenced by the electrified calciner, the heat pump, and the CO₂ purification and compression processes, to 28% and 26% for the Ref +MEA and OC-HK cases, respectively. CAPEX plays an important role in all the technologies, ranging from 30% to 20% of the overall CAC, with the MEA system being the most capex-intensive alternative, mainly impacted by the cost of the heat pump. This is a significant aspect of the economic evaluation to emphasize, given the high uncertainty regarding the CAPEX of the new technologies. Finally, 21-24% of the CAC corresponds to T&S of the captured CO₂ in the partially electrified technologies and the benchmark, with the highest share obtained for the OC-HK case. Whereas in the eC-pK alternative, the share is only 13%. In this case, the high electrification degree of the process removes completely the direct fuel-related CO2 emissions, reducing the amount of captured CO₂ to be managed.

It is also worth explaining the reasons for the values obtained for the reference plant coupled with a MEA system, which are higher than previous ones reported in the literature. For example, the estimated CAC in the framework of the CEMCAP project is 80 €/t_{CO2} (Gardarsdottir et al., 2019). The significant difference can be explained with the year in which the capital investment is made, the technology for steam generation, and the addition of T&S costs. The total plant cost (TPC), originally estimated for the year 2014, has been adjusted to 2022 values using the CEPCI index. This meant a 42% increase in TPC. Second, instead of a natural gas boiler, in this study we assumed that the heat for the regeneration unit is supplied with a heat pump, which has a larger investment cost as well as lower maturity, reflected in higher contingency. Likewise, the contributions to the OPEX are changed, from an unabated natural gas-based heat generation system to an electrified one. Finally, the cost for T&S had not been previously included, adding another \notin 25 per captured CO₂.

There are parameters of particular relevance for the economic assessment of strategies for the decarbonization of the cement industry through electrification: the cost of electricity, the cost of T&S, and the cost of CO_2 emissions. To better understand the relationship between these factors and the economic performances of the electrified alternatives, two sensitivity analyses are conducted.

Fig. 13 shows the results from the sensitivity analysis on the electricity price versus the additional cost of clinker. The solid lines represent the trend under baseline conditions, with a T&S cost of $25 \text{ } \text{ } \text{/t}_{CO2}$,



Fig. 13. Sensitivity analysis on the electricity price. For a given case, the solid lines represent the trend under baseline conditions, with a T&S cost of 25 $\ell/t_{\rm CO2}$, while the shaded area between the dashed lines is the result of varying the T&S cost in the range of 10–50 $\ell/t_{\rm CO2}$.

while the shaded area within the dashed lines is the result of varying the T&S cost in the range of 10–50 $\ell/t_{\rm CO2}$. Therefore, the width of the shaded area indicates the impact of T&S on the economic performance.

As expected, the alternative with the largest dependence on the electricity price is eC-pK, which entails a complete electrification of the conventional manufacturing process. Next, is the eC-afK with the partial electrification of the pre-calciner operation, followed by the Ref + MEA and the OC-HK cases. These last two cases follow similar trends between each other, with the semi-electrified alternative outperforming the benchmark through the whole range of electricity prices (maintaining the same cost of T&S). Furthermore, it is worth noting that all three electrified cases have a lower impact on the additional COC in regions with high availability of low-cost electricity, outperforming the benchmark MEA system. In particular, the crossover point for the eC-pK and the eC-afK cases is found at the electricity price of 33 and 119 \notin /MWh, respectively.

Finally, the impact to the additional COC from varying the cost of CO_2 emissions is represented in Fig. 14. In addition to the performance of the Ref + MEA system and the electrified cases, the additional COC from the CO₂ emissions of the reference cement plant without CO₂ mitigation is included. The figure underscore how the rising price of CO₂ emissions, expected to affect the cement industry in the near future, influences the economic feasibility of different technologies when compared against the conventional process. The eC-afK case is the best performing between the electrified alternatives below a carbon tax of 100 \notin /t_{CO2}, closely followed by the OC-HK case. Both intersect with the reference plant at a carbon tax of approximately 100 €/t_{CO2}. On the other hand, the eC-pK case starts to outcompete the reference case at a carbon tax above 139 ℓ/t_{CO2} , however with a steeper slope than the benchmark. Therefore, it will always underperform the Ref + MEA system under the assessed conditions. Conversely, the OC-HK case has a descending slope, meaning that the technology benefits from an increase in the cost of CO₂ emissions, being a net negative emission process.

3.3. Electrification and carbon capture

To understand the role of electrification in the effort of decarbonizing the cement industry, it is important to compare its performance against the use of carbon capture technologies. To this end, two comparative analysis are presented below, i) the economic performance of an electrified cement plant with and without CO₂ capture, and ii) the techno-economic comparison of the electrified cement plants versus a cement plant with first generation oxyfuel carbon capture.

To illustrate the need of carbon capture, regardless of the electrification degree, Fig. 15 compares the additional COC of the eC-pK case with and without the CPU, assuming a carbon tax of 100 ℓ /t_{CO2} and T&S costs of 25 ℓ /t_{CO2}. Without carbon capture, the added value of the CAPEX and the fixed costs is reduced by 35%, yet the additional COC increases 23% because of the penalty of emitting the CO₂. Even though



Fig. 14. Sensitivity analysis on the cost of CO₂ emissions.

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Fig. 15. Comparison of the additional COC of the eC-pK case with and without CO_2 capture.

the fully electrified case completely removes the fuel-related $\rm CO_2$ emissions, around 67% of the overall $\rm CO_2$ from the reference cement plant are still released (process emissions). If no carbon capture technologies are considered in the assessment, the cost of emitting this $\rm CO_2$ will exceed the investment in additional equipment.

Table 3 shows a comparison of the techno-economic KPIs between the electrified cement plants and a cement plant with carbon capture based on oxyfuel technology. The KPIs for the oxyfuel plant were developed from the CEMCAP project (Gardarsdottir et al., 2019), updating the price and carbon intensity of the electricity to 50 €/MWh and 50 kg_{CO2}/MWh, respectively, as well as adding 100 €/t_{CO2} for unabated CO₂ and 25 €/t_{CO2} for T&S of the captured CO₂, along with adjusting the TPC to 2022 values. According to the results, the eC-pK, OC-HK, and eC-afK cases demonstrate a 73%, 30%, and 30% additional COC increase compared to oxyfuel technology, respectively. Likewise, the CAC is increased 83% in the fully electrified case and 32% in the partially electrified alternatives, with respect to the carbon capture case. A sensitivity analysis on the electricity price indicates that the breakeven points for the additional COC of the eC-pK and the eC-afK cases versus oxyfuel technology is found at 3.7 and 19.0 €/MWh, respectively. In contrast, the OC-HK case becomes competitive only at negative prices below −13 €/MWh.

The breakdown of the additional COC is presented in Fig. 16. The oxyfuel technology is also heavily influenced by the capital investment, which includes a CPU, ASU, and modifications to the rotary kiln (to reduce false air ingress) and the clinker cooler (to operate with two sections, one with air and the other with a mixture of O₂ and recirculated CO₂). But unlike the electrified alternatives, it is less dependent on the electricity price. It is also worth noting that the cost of T&S represents 25% of the cost of emitting CO₂. Therefore, the economic performance can be improved further by assuming a higher capture rate, instead of 90%. For every additional 1% of capture rate, the additional COC could be reduced 0.66 ϵ/t_{clk} .

4. Conclusions

In this work, a comparative assessment of four alternatives for the decarbonization of the cement industry through electrification is

Table 3

Comparison of techno-economic KPIs between electrified alternatives and oxyfuel technology.

Techno-economic KPI		eC-pK	OC-HK	eC-afK	Oxyfuel
Capture rate	-	92%	95%	99%	90%
Specific CO ₂ emissions	kg _{CO2} /t _{clk}	110.9	-15.7	14.1	102.4
CAC	€/t _{CO2}	138.8	100.1	100.5	76.0
Additional COC	€/t _{clk}	116.0	86.7	87.0	67.2
Breakeven electricity price	€/MWh	3.7	-13.0	19.0	_



Fig. 16. Comparison of the additional COC between the electrified alternatives and the oxyfuel technology.

conducted based on process simulation using Aspen Plus and the estimation of techno-economic KPIs. The four cases included in the assessment are: (i) eC-pK: electrified pre-calciner coupled with plasma generators in the rotary kiln; (ii) OC-HK: oxy-combustion of alternative fuels in the pre-calciner and combustion of high-temperature electrolytic H₂ in the rotary kiln; (iii) eC-afK: electrified pre-calciner and combustion of alternative fuels in the rotary kiln; (iii) eC-afK: electrified pre-calciner and combustion of alternative fuels in the rotary kiln, with post-combustion CO_2 capture (PCC); and (iv) e-hCHS: electrification of the hydraulic Calcium Hydro Silicate (hCHS) production process including the supply of hydrated lime. The results were compared against a reference cement plant without CO_2 capture, a reference plant with a PCC system based on MEA solvent and regeneration via heat pump, and a reference hCHS production process, used as the benchmarks.

The following main conclusions can be listed:

- The increase in electricity demand from the electrified alternatives varies depending on the technology. The eC-pK case shows the highest increase in electricity consumption (1,341 kWh/t_{clk}), followed by e-hCHS (1,245 kWh/t_{clk}), eC-afK (929 kWh/t_{clk}), and finally OC-HK (598 kWh/t_{clk}). Processes with higher electricity consumption result in lower fuel consumption, as the total energy input (electricity + fuel) varies in a relatively narrow range of 4.5–5.1 GJ/t_{clk}.
- All the assessed cases achieve very low direct CO2 emissions, thanks to both electrification and high CO₂ capture efficiencies (95-100%) of the residual process and fuel CO₂. Cases burning alternative fuels achieve the highest CO₂ avoidance rate and lowest net emissions, thanks to the credits associated to the capture of biogenic CO₂. The OC-HK case shows the best performance in terms of direct and equivalent CO2 emissions avoided (101.8% of equivalent CO2 emissions), because of the biogenic emissions present in the alternative fuel and the high capture rate. Nevertheless, this alternative also has to deal with the largest amount of captured CO₂ (835 kg_{CO2}/ton_{clk}), and the additional economic and environmental burden of the transport & storage (T&S) infrastructure. The eC-pK and e-hCHS cases have a lower ratio of net CO2 emissions avoided (87.2% and 92.8%, respectively), but also lower amount of captured CO₂ to be managed: 536 and 357 kg_{CO2}/ton_{clk}, respectively. Between these ranges, the eC-afK case achieves 98.4% of equivalent CO2 emissions avoided and 707 kg_{CO2}/ton_{clk} of captured CO₂ to be managed in the T&S infrastructure. Overall, a trade-off can be observed between a higher electrification of the cement manufacturing process and the amount of captured CO_2 to be managed.
- The carbon intensity of the grid plays a major role in the environmental performance of the technologies. The results show that in a low-carbon power grid dominated by renewables/nuclear sources, all the electrified cases can achieve a considerable reduction in the carbon footprint of cement production. In the carbon intensity range

of 0–50 kg/MWh, the $\rm CO_2$ emissions from the electrified alternatives vary between:

 $\begin{array}{l} \circ \text{Ref} + \text{MEA: } 43\text{-}77 \ kg_{\text{CO2}}/t_{\text{clk}}; \\ \circ \text{eC-pK: } 44\text{-}111 \ kg_{\text{CO2}}/t_{\text{clk}}; \\ \circ \text{OC-HK: } (-46) - (-16) \ kg_{\text{CO2}}/t_{\text{clk}}; \\ \circ \text{eC-afK: } (-32) - 14 \ kg_{\text{CO2}}/t_{\text{clk}}; \\ \circ \text{e-hCHS: } 0\text{-}62 \ kg_{\text{CO2}}/t_{\text{clk}}; \\ \end{array}$

If the carbon intensity of the power grid is between 330 and 400 kg/ MWh, representative of a Natural Gas Combined Cycle plant, the CO_2 emissions range between:

 $\begin{array}{l} \circ \text{Ref} + \text{MEA: } 266 - 313 \ kg_{\text{CO2}}/t_{clk}; \\ \circ \text{eC-pK: } 486 - 580 \ kg_{\text{CO2}}/t_{clk}; \\ \circ \text{OC-HK: } 152 - 194 \ kg_{\text{CO2}}/t_{clk}; \\ \circ \text{eC-afK: } 274 - 339 \ kg_{\text{CO2}}/t_{clk}; \\ \circ \text{e-hCHS: } 411 - 498 \ kg_{\text{CO2}}/t_{clk}; \\ \end{array}$

- Under the baseline conditions of the economic assessment (electricity cost: 50 ℓ /MWh, cost of CO₂ T&S: 25 ℓ /t_{CO2}), the additional cost of clinker (COC) with respect to benchmark plant with zero CO₂ emission cost and the cost of CO₂ avoided (CAC) for all the technologies are:

$$\begin{split} & \circ \text{Benchmark Ref} + \text{MEA case: } 104.4 \ \texttt{\&ft}_{clk} \ \text{and } 122.7 \ \texttt{\&ft}_{CO2}; \\ & \circ \text{eC-pK case: } 115.9 \ \texttt{\&ft}_{clk} \ \text{and } 138.8 \ \texttt{\&ft}_{CO2}; \\ & \circ \text{OC-HK case: } 86.7 \ \texttt{\&ft}_{clk} \ \text{and } 100.1 \ \texttt{\&ft}_{CO2}; \\ & \circ \text{eC-afK case: } 87.0 \ \texttt{\&ft}_{clk} \ \text{and } 100.5 \ \texttt{\&ft}_{CO2}. \end{split}$$

- The economic performance of the electrified alternatives is highly dependent on the electricity price. Therefore, a sensitivity analysis is conducted to understand the impact of changing the electricity price over the additional COC. The results showed that the OC-HK case outperforms the reference technology with MEA-based CO₂ capture independently of the cost of electricity. The eC-afK case outperforms the reference MEA-based plant up to an electricity price of 119 \notin /MWh, but has a higher COC than the OC-HK case, except for very low electricity price (<19 \notin /MWh). Finally, the eC-pK case is more competitive than the reference MEA case only for electricity prices below 32 \notin /MWh.
- The role of electrification in the decarbonization of the cement industry is linked to the use of carbon capture and storage (CCS). For example, without carbon capture, the emission reduction of the eCpK case would become only 33% (vs. 87.2%) and the additional COC increases by 23% because of the emission of unabated processrelated CO₂. Furthermore, a comparison between the electrified alternatives and a cement plant with oxyfuel technology showed that the cost of implementing CCS strategies is economically more competitive than electrification, unless extremely low-cost electricity is available (<3.7 €/MWh for the eC-pK case, or < 19 €/MWh for the eC-afK case).

CRediT authorship contribution statement

Sebastian Quevedo Parra: Conceptualization, Methodology, Investigation, Formal analysis, Software, Visualization, Writing – original draft. **Matteo C. Romano:** Conceptualization, Methodology, Supervision, Validation, Writing – review & editing, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Matteo C. Romano reports financial support was provided by Innovandi Global Cement and Concrete Research Network. Matteo C. Romano reports a relationship with Innovandi Global Cement and Concrete Research Network that includes: funding grants.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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