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Retrofitting partial oxyfuel and Integrated Ca-Looping technologies to an existing cement plant: a case study

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Abstract

The present document describes the potential retrofit of an existing cement plant with carbon capture technologies applied in two sequential steps. The pathway proposed consists in a first retrofit through partial oxyfuel followed by the integrated calcium looping (CaL) technology. This kind of applications may represent a promising strategy for the decarbonization route in the cement sector without introducing chemical solvents or special components, in particular for existing cement kilns that may need to be revamped. The cement plant selected for this study is the 0.5 Mt_{cem}/y Colleferro facility owned by Italcementi-HeidelbergCement. This study analyses the mass & energy balances of the partial oxyfuel, and the integrated CaL process retrofitted to the existing cement plant. The results of the two CCS technologies are then compared in terms of CO_2 emission reduction and energy consumption with the reference plant without CO_2 capture. The scope of this analysis is to evaluate the impact of carbon capture technologies on the cement production process. The process simulation software Aspen Plus V10.0® has been employed to develop the model for the three different plant configurations (i.e., the base case w/o carbon capture, the partial oxyfuel mode, and the integrated CaL). The base case has been validated using field measurements coming directly from the Colleferro plant. From this process flow model, the two CCS technologies have been developed according to the specific process requirements. Results show that a maximum reduction in CO_2 emissions of 92.4% is possible with the integrated CaL, while the partial oxyfuel enables to capture 71.7% of the CO_2 generated in the plant.

Keywords: CO_2 capture; Calcium Looping ; Colleferro Cement Plant; Retrofit; Partial Oxyfuel; Integrated CaL

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1. Introduction

The present work has been developed in the framework of the CLEANKER project [1], focused on the experimental validation and on the conceptual scale-up of the integrated Calcium Looping system (CaL), under demonstration at TRL7 in the pilot plant in the Vernasca cement plant (owned by Buzzi Unicem).

The state-of-the-art cement production process results in direct CO_2 emissions of nearly 860 kg_{CO2} per tonne of clinker [2], making this sector responsible for 8% of the total anthropogenic CO_2 emission worldwide [3].

Carbon capture & storage is considered by Cembureau the solution with the highest potential impact in reducing CO_2 emissions from the cement value chain [4].

The case study here envisaged refers to the existing cement plant located in Colleferro (Italy), owned by Italcementi (HeidelbergCement group). The Colleferro facility, schematized in Fig. 2, has a production capacity of about 0.5 Mt_{cem}/y and is characterized by a four-stage preheater without precalciner, an auxiliary firing at kiln back end, a 67 m-long rotary kiln and a clinker cooler.

The pathway analysed in the case study is based on adopting the partial oxyfuel concept to the Colleferro facility, which can be subsequently followed by the implementation of the integrated CaL concept [5].

The first step requires the assessment of the partial oxyfuel configuration, via the installation of an oxy-fired, entrained flow (EF) calciner, an Air Separation Unit (ASU), a CO_2 purification unit (CPU) and a heat recovery power plant. As in the case of conventional revamping with an air-fired calciner, the operation allows increasing the cement plant productivity by around 40%, thanks to the additional fuel input in the new calciner. Given that raw meal calcination is the most carbon intensive process in cement making, the oxyfuel calciner allows avoiding around 75% of the total carbon dioxide generated from the raw meal calcination and fuel combustion.

In the second retrofit step, the integrated CaL configuration is assessed via the installation of an EF carbonator. The EF carbonator is designed to capture from 60% up to 90% of the residual CO_2 from the rotary kiln exhausts by means of the carbonation reaction. Decarbonized effluents can be sent to the heat recovery section and then to the conditioning/mill section of the cement plant, as in the partial oxyfuel configuration.

In both the two carbon capture processes analysed (conceptually schematized in Fig. 1), the raw meal preheating is carried out in the existing four-stage tower following a hybrid set up: three cyclone stages are supplied with the CO_2 rich gas from the oxy-calciner, while the other one receives the kiln off gas.

The partial oxyfuel and integrated CaL options with the hybrid preheater, could be retrofitted to most of the existing cement plants configurations, and represent a valid alternative to both the dominant solvent-based CO_2 capture processes (penalized by the significant additional thermal energy required for the solvent regeneration) and to the full oxyfuel configuration (tight sealing against “false air” in-leakages in the rotary kiln and the clinker cooler need to be proven).

Mass and energy balances of the various cement plant configurations, for both the baseline case (w/o carbon capture) and the two cases equipped with CO_2 capture systems, have been calculated through process simulations in Aspen Plus V10.0® including a 1D model of the EF carbonator, and calibrated with real plant data from the Colleferro cement plant.

The paper begins with an overview in section 2 of the methodology employed in Aspen Plus to simulate the three configurations analyzed. Section 3 presents and describes the validation of the Aspen Plus process simulation and the main results obtained. Finally, section 4 summarizes the work done and the outcomes, with some considerations on future prospects.

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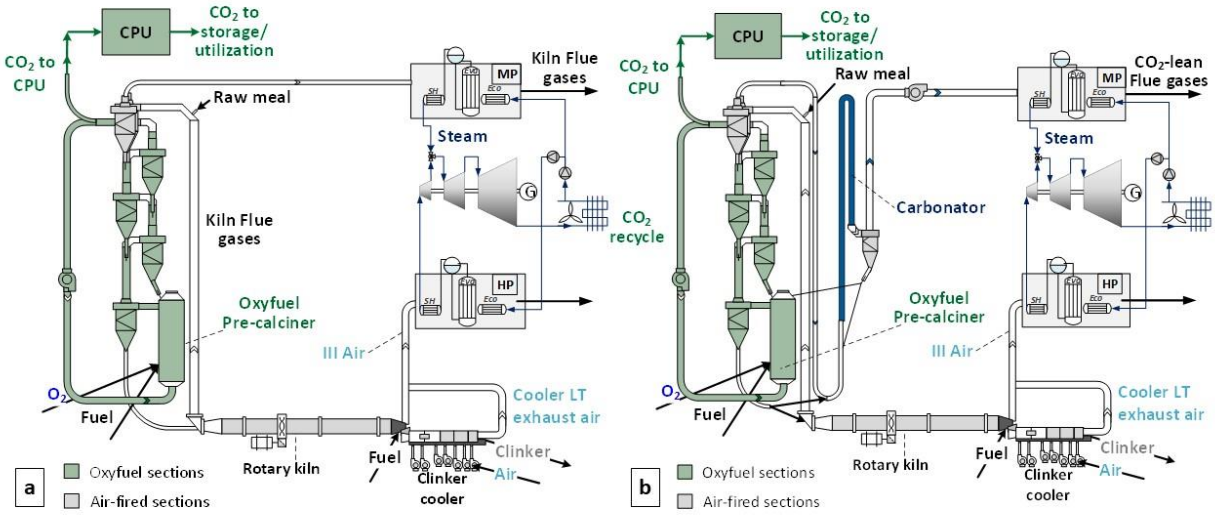

Fig. 1: Conceptual scheme of (a) the partial oxyfuel, and (b) integrated CaL technology, both applied to the Colleferro cement plant with the hybrid preheater configuration and the heat recovery steam cycle.

2. Process model

In order to evaluate the technical KPIs of the two CCS technologies proposed, a detailed process design & simulation has been carried out. The models for the three configurations (i.e., the base case, partial oxyfuel mode, and the integrated CaL), have been developed using the Aspen Plus V10.0® process simulation software.

To model the gas phase in the process simulation, the databases available in Aspen Plus for the gaseous species and the Peng-Robinson equation of state have been adopted to calculate thermodynamic properties. For the pure components thermophysical properties of the solid species (specific heat capacity, heat of formation, etc.), the databanks provided by Barin [6] and Matschei [7] have been implemented into Aspen Plus.

The baseline case (i.e., the Colleferro cement plant in its current configuration w/o carbon capture systems) was implemented and simulated in Aspen Plus. The process model has been calibrated with real data from the plant. The validated reference case served as starting point for the development of the partial oxyfuel and integrated CaL configurations. The new equipment for CO_2 capture has been added in the AspenPlus model, and the process flowsheet modified according to the technical requirements of the two CCS processes. In line with a retrofitting logic, the cement plant configuration and the CO_2 capture process parameters have been tuned to avoid significant modifications in the existing plant (mainly related with the preheating tower).

In both CCS applications, the CO_2-rich stream is further processed in the CPU, where a high purity (> 95%vol.) CO_2 stream is obtained at 110 bar, meeting the safety and techno-economic standards of the transportation, utilization, or storage systems [8].

2.1. Reference cement plant

The baseline configuration of the Colleferro cement plant is reported in Fig. 2. The facility is a small size cement kiln (cement production capacity of 0.5 Mt_{cem}/y), without a pre-calcination section, with an auxiliary firing system at the rotary kiln back end that shares 21% of the total heat input.

The raw meal chemical composition and feeding flow rate have been defined in the process model according to measurements taken directly in the Colleferro cement plant.

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By contrast, the total fuel flow rate has been adjusted in the simulations to provide the required energy input for the reaction of formation and the melting process of the clinker compounds. In this way, it is possible to check if the energy consumption estimated by the Aspen Plus model leads to results that are in line with the real plant performances. In particular, the amount of fuel input to the rotary kiln is varied to achieve a clinker temperature at kiln discharge of 1380°C (average measured value inside the plant).

The preheating tower of the Colleferro cement plant is composed of four cyclone-stages. In the Aspen simulation, each cyclone is evaluated with the embedded cyclone model. The required input parameters are the geometric dimensions of the cyclone, which have been estimated from technical drawings, and wall friction coefficient. From the inlet gas flow rate, the Aspen model calculates the cyclone efficiency using the Muschelknautz method [9].

The total pressure drop along the preheater is estimated as the contribution of two terms: the first one is given by the cyclone pressure drop calculated by the Muschelknautz model, while the second term accounts for the pressure drops due to gas-solid acceleration, gas-solid wall friction, and gas-solid static head, according to the methodology outlined in Rhodes [10].

The clinker cooler is modelled using the information available from the cement plant process data. The mass flow rate of secondary air is controlled to achieve the measured oxygen concentration in the kiln back end gas. The vent air is obtained from the closure of the energy balance, with the given the clinker outlet temperature.

Heat losses from the main process units (kiln, cooler and preheater) have been provided by mass and energy balance calculations on the Colleferro plant and are given as input parameters to the Aspen Plus model.

The false air inlet in the rotary kiln and in the clinker cooler are taken from the measurements carried out in the plant. On the contrary, the false air flow in the preheating tower is adjusted to match the oxygen concentration profile along the tower.

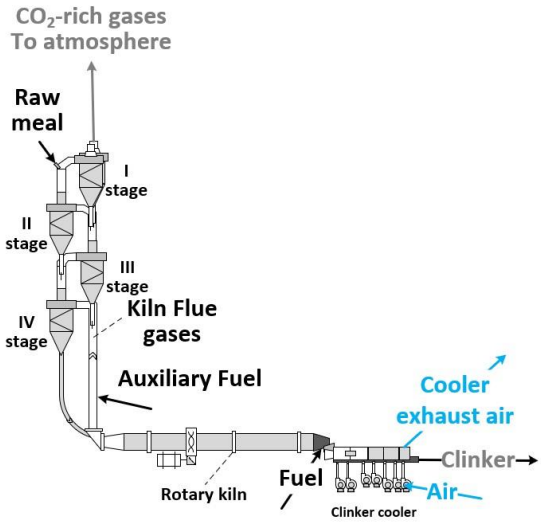

Fig. 2: Conceptual scheme of the Colleferro cement plant in the current configuration.

2.2. CCS technology retrofit: Partial oxyfuel application

The operating parameters for the oxy-fired calciner were retrieved from the simulations carried out by VDZ on the best available technology (BAT 3000) for the cement process [2]. The calcination degree of the solid material exiting the reactor (defined as the ratio between moles of Ca reacted vs total moles of Ca) has been set to 85%. The outlet temperature of the gas and the oxygen concentration at calciner outlet have been set equal to 920°C and 2.621%, respectively. The latter is a good representation of the outlet condition of a conventional precalciner section, while the

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outlet temperature value should guarantee the targeted calcination degree in a higher CO_2 rich atmosphere due to oxyfuel condition.

The oxygen concentration and the calciner outlet temperature are respectively controlled by the amount of fuel and the flow rate of oxygen from the ASU fed to the calciner burner. In addition, a recirculation duct for the CO_2-rich flue gas from the top of the preheating tower back to the calciner has been added. This component enables to control the flue gas velocity in the preheater, and therefore the cyclone efficiency calculated with the Muschelknautz method. The amount of flue gas recirculated is adjusted in order to maintain the same gas velocity as in the reference case. According to this approach, the partial oxyfuel technology could be retrofitted to the Colleferro cement plant without major modifications to the preheating tower.

On the contrary, a revamping or, most likely, a full replacement of the clinker cooler section with a new unit will be necessary. Indeed, the increased productivity of the cement plant will require a higher flow rate of cooling air. The design of the clinker cooler section in Aspen Plus has been carried out with the same logic employed for the baseline case. In this case a tertiary air stream is added, and the clinker outlet temperature is set to 115°C in line with state of the art configurations. Since no tertiary air can be fed to the oxyfuel calciner (to avoid CO_2 dilution), the excess air can be exploited in a heat recovery section, together with the flue gases exiting the rotary kiln. This solution is displayed in the schemes of Fig. 1.

The Aspen model for the CPU developed in the work of Magli et al. [11] has been implemented in this study to evaluate the CO_2 purity and the energy consumption of this unit. For the ASU, a specific power consumption of 230 kWh_e/t_{O2} is considered, as reported in De Lena et al. [5].

No detailed considerations are done for the thermal power recovery section and the integration with a power cycle, but the potential energy savings of exploiting the residual heat content in the flue gases exiting the process are estimated.

2.3. CCS technology retrofit: Integrated CaL technology

From the partial oxyfuel configuration, the integrated CaL process is built in Aspen Plus via the addition of the entrained flow carbonator, as depicted in Fig. 1.

In real applications the carbonator is long transport pipe designed to capture a targeted amount of CO_2 from the flue gases by means of the carbonation reaction. In the process simulation framework, this component is modelled as an adiabatic reactor using the 1D model developed by Spinelli et al. [12]. Specifically, this model receives in input the composition of the inlet gas/solid mixture and the temperature resulting from the Aspen plus simulation. As output, it calculates the CO_2 capture efficiency, and the carbonator outlet gas/solid composition.

In this study, the installation of the sorbent cooler to cool down the temperature of the flow at carbonator inlet is not covered due to the hybrid set up of the preheating tower. Indeed, a gas/solid mixing temperature of around 600°C is achieved at the carbonator inlet by exploiting some of the residual heat in the kiln effluents for preheating the raw meal in the first cyclone stage. Another advantage related to the hybrid preheater, and valid also for the partial oxyfuel process, is that the first cyclone stage of the preheater serves to clean the kiln flue gases from sticky dust and corrosive compounds. In the context of heat recovery from the flue gases, this can be of relevant importance to avoid risks of corrosion and clogging.

3. Aspen Plus simulation results

The results presented in this section refer to the mass and energy balance outcomes of the Aspen Plus simulations for the reference base case (i.e., the Colleferro cement plant w/o carbon capture), and the two CCS technologies with the preheating tower in the hybrid setup.

3.1. Validation of the Aspen Plus simulation with Colleferro plant data

In order to provide a solid reference case from which the two CCS configurations can be evaluated in detail, a consistent process model has to be developed. This has been done within the Aspen Plus process simulation

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framework, where mass and energy balances of the reference cement plant without CO_2 capture have been evaluated. The results of the model validation are shown in Table 1.

The deviation between the model results and the plant data is useful to provide an indication of the accuracy and reliability of the Aspen Plus model. A relevant difference between plant data and the Aspen Plus simulation is represented by the fuel consumption and the specific CO_2 emission. The relative error is indeed in the order of 6% for the two parameters. Another significant deviation occurs in the gas flow rate at the rotary kiln outlet, which are overestimated by about 24.5%.

Table 1. Main results of the Aspen Plus simulation for the baseline case compared with real plant data.

	Colleferro plant data	Aspen simulation
Clinker production [tpd]	1252	1260
Fuel input to rotary kiln [t/h]	5.59	5.84
Specific fuel consumption [MJ/tclk]	3548	3680
Rotary kiln gas outlet flow rate [Nm^3/h]	51227	63778
Flue gas flow rate from preheater [Nm^3/h]	83988	85277
Flue gas preheater outlet composition - O_2 [% mol,dry]	4.6	4.6
Flue gas preheater outlet composition - CO_2 [% mol,dry]	29.7	29.5
Flue gas IV outlet composition - O_2 [% mol,dry]	2.35	2.36
Flue gas IV outlet composition - CO_2 [% mol,dry]	33.40	33.39
I stage outlet gas velocity [m/s]	14.14	14.60
II stage outlet gas velocity [m/s]	21.86	22.81
III stage outlet gas velocity [m/s]	15.47	15.41
IV stage outlet gas velocity [m/s]	15.72	15.85
CO_2 specific emission [kg_{CO2}/tclk]	859	905

3.2. Model results for the CCS technologies

Some of the main outcomes of this work have been collected in Table 2, which reports the most significant results of the mass and energy balances for the three process configurations: (i) the Colleferro cement plant w/o CO_2 capture, (ii) the Colleferro cement plant in the partial oxyfuel configuration, and (iii) the integrated CaL process applied to the Colleferro facility.

The CO_2 content in the flue gases downstream the conditioning line is reduced from 12.21% in the baseline cement plant. to 3.05% and to 0.7% in the partial oxyfuel and integrated CaL process, respectively. The clinker production increases by approximately 40% if one of the CCS solutions is adopted, due to the installation of the pre-calciner section. These retrofitted plants require a significant amount of fuel input to treat the additional raw material fed to the plant and to sustain the CO_2 capture unit. Indeed, the specific fuel consumption increases from a minimum of 3680 MJ/tclk in the base case, to a maximum of 4506 MJ/tclk in the integrated CaL configuration. This not negligible increase of primary energy consumption can be partially compensated by heat recovery from the flue gas stream exiting the plant. Indeed, in the partial oxyfuel configuration a specific electric power of 78 kWh/tclk can be recovered from a power cycle, while in the integrated CaL process a potential saving of 113.15 kWh/tclk is calculated. These values have been estimated by considering the heat available above 250°C from the hot gas streams (i.e., CO_2-rich gas from the preheater, tertiary air, and carbonator effluents), and a gross electric efficiency of 29.1% for the heat recovery steam cycle[2].

As can be noticed from Table 2, there is a good agreement in the three different configurations, between the separation efficiencies of the cyclones, and thus the gas velocity in the preheating tower along the II, III and IV stage. On the contrary, a relevant deviation is reported for the I stage of the preheater tower. This is mainly due to the

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significantly lower gas flow rate from the rotary kiln, both in partial oxyfuel and integrated CaL applications, which is sent to the top cyclone stage in the two CCS configurations analyzed.

In the context of a retrofit application, this can be partially solved by adopting just one of the two cyclones present in the top stage. In this way, for the same gas volumetric flow rate, the inlet velocity increases thanks to the cross section area reduction. If this solution would be adopted, the new gas velocities calculated are comparable with the reference ones, as indicated by the results listed in Table 2 for the I stage.

Another operation that will probably need to be undertaken after the installation of CO_2 capture technologies consists in the maintenance of the preheating tower to minimize the false air inlets and avoid dilution of the CO_2-rich gas from the oxy-calciner. The results reported in Table 2 feature the case in which the false air inlets have been reduced by 90% compared to the reference plant model during the simulation of the partial oxyfuel and integrated CaL technologies. This results in a relatively high CO_2 concentration in the gas stream from the pre-calciner section that has to be treated in the CPU (around 90%mol on a dry basis). Therefore, the CO_2 recovery in this process unit reaches values of 97%, and the direct CO_2 emissions are reduced respectively to 71.7% and 92.4% in the partial oxyfuel and in the integrated CaL process (in this latter case the CO_2 vented from the CPI is recycled to the carbonator).

Table 2. Comparison of the main process parameters for the three different assessed processes.

	Base case	Partial oxyfuel	Integrated CaL
Clinker production [tpd]	1260	1751	1752
Fuel input to rotary kiln [t/h] , [t/tclk]	5.84, 0.11	3.28, 0.045	3.29, 0.045
Fuel input to oxy-calciner [t/h]	-	6.66, 0.091	8.45, 0.116
Total fuel consumption [t/h]	5.84	9.94	11.74
Specific fuel consumption [MJ/tclk]	3680	4506	5318
Rotary kiln gas outlet flow rate [Nm^3/h], [Nm^3/kgclk]	63778, 1.21	35301, 0.48	35359, 0.48
Rotary kiln gas outlet temperature [°C]	1112	1109	1108
Flue gas flow rate from preheater [Nm^3/h], [Nm^3/kgclk]	85277, 1.62	36048, 0.49	32610, 0.45
Flue gas preheater outlet composition - O_2 [% mol,dry]	4.63%	1.90%	4.41%*
Flue gas preheater outlet composition - CO_2 [% mol,dry]	29.67%	25.12%	8.34%*
O_2 content in the oxidizing gas to the calciner [% mol,dry]	-	24.85%	34.41%
Cyclone efficiency (1^{st}/2^{nd}/3^{rd}/4^{th} stage) [%]	91.9/80.0/77.0/75.4	95.5/80.6/75.5/77.2	95.7/81.5/77.5/85.3
Rotary kiln outlet gas velocity [m/s]	9.1	4.9	4.9
I stage outlet gas velocity [m/s]	14.60	13.2	12.9
II stage outlet gas velocity [m/s]	22.81	23.2	22.9
III stage outlet gas velocity [m/s]	15.41	15.8	15.8
IV stage outlet gas velocity [m/s]	15.85	17.4	17.6
CO_2-rich gas to CPU [kg/s]	-	14.59	19.85
CO_2 content to CPU [%mol,dry]	-	89.12%	90.58%
CO_2 vented from CPU [kg/s]	-	0.56	0.55
CPU CO_2 recovery [%]	-	96.15%	97.24%
CPU electric consumption [kW], [kWh/tclk]	-	5865.4, 80.4	8196.8, 112.3
CPU electric consumption [kJ/kgCO_2]	-	418.92	423.14
CO_2 purity [% mol,dry]	-	97.16%	96.36%
Oxygen from ASU [t/h]	-	18.73	23.62
ASU electric consumption [kW], [kWh/tclk]	-	4307.2, 59.0	5432.8, 74.4
Heat available for heat recovery** [kW]	-	19560	33403
Electric power production [MWe]	-	5692	9720

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Electricity consumption [kWh/tclk]	51.9	113.3	106.7
Electricity demand increase [%]	-	118%	105%
Direct CO_2 emission factor [$kgCO_2$/tclk]	905.29	256.14	69.04
Direct CO_2 emission reduction [%]	-	71.7%	92.4%

*downstream the carbonator

**above 250°C from the flue gases exiting the process

4. Conclusion

Carbon capture technologies are expected to play a crucial role for the decarbonization of hard-to-abate sectors like the cement industry.

This work focuses on the retrofitting of two CCS applications to an existing cement plant located in Colleferro (Italy) owned by Italcementi (HeidelbergCement Group). Their impact is analyzed in terms of CO_2 emission reduction and energy performances. In order to properly evaluate mass and energy balances, a detailed model of the different process configuration has been developed in Aspen Plus V10.0®.

At first, the process model for the base case (i.e., the cement plant without CO_2 capture) was created and the simulation validated with field data directly from the plant. The results indicate that the Aspen Plus model can reproduce with a reasonable accuracy the inlet/outlet fluxes and the energy consumption of the Colleferro cement plant.

Once the cement plant model was calibrated, the two CCS technologies have been implemented in the Aspen Plus simulation. The two technologies enable a drastic reduction of CO_2 emissions. For the partial oxyfuel application, the direct CO_2 emission are cut by 71.7% while for the integrated CaL, the reduction is of 92.4%.

The fuel consumption increases by 70% for the partial oxyfuel and by almost 101% if the integrated CaL technology is adopted. This corresponds to an increase in the primary energy consumption for producing 1 tonne of clinker equal to 22.4% and 44.5% for the partial oxyfuel and integrated CaL respectively. The incremental difference between these two cases is due to the fact that in the integrated CaL additional energy is required to regenerate the CO_2 sorbent.

Another penalization in terms of energy consumption is the increase in the electricity demand due to the installation of the ASU for oxygen requirement and the CPU for the CO_2 purification (+118% for partial oxyfuel and +105% for integrated CaL).

This study suggests the technical feasibility of retrofitting in two subsequent steps the partial oxyfuel and the integrated CaL technology. Indeed, the results of the Aspen Plus simulations show how these two solutions can be implemented without major modifications to ducts and cyclones, since the gas velocities in the preheater are comparable to the values of the baseline operation thanks to the control on the CO_2 recycle.

Future works should address a more detailed thermal integration with the heat recovery steam cycle to properly account the indirect (Scope 2) CO_2 emissions and should rely on CaL reactors validated with the experimental data from the CLEANKER test campaigns. In addition, an economic analysis regarding CAPEX (construction works and plant modifications) and OPEX (additional fuel and electricity consumption) of the presented retrofit study, will enable to evaluate the effect of the installation of these CO_2 capture technologies on the cost of clinker.

Acknowledgements

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Nomenclature

1D	One dimensional
ASU	Air Separation Unit
BAT	Best Available Technologies
CaL	Calcium Looping

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CCS	Carbon Capture & Storage
CPU	CO_2 Purification Unit
EF	Entrained Flow
GHG	Green House Gases
KPI	Key Performance Indicators
tclk	Metric tonnes of clinker
t_{cem}	Metric tonnes of cement
tpd	Metric tonnes per day
%mol,dry	Molar fraction on a dry basis

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