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## CO<sub>2</sub> capture in cement plants by “Tail-End” Calcium Looping process

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### Abstract

In this work the integration of the Calcium-Looping (CaL) process, used as a post-combustion CO<sub>2</sub> capture system, into a cement kiln was analyzed by means of process simulations. The results show that capture efficiencies of about 90% can be achieved with operating conditions of CaL reactors similar to those for power generation applications. The integration of the CaL process increases the fuel consumption of the cement kiln, but the additional primary energy introduced for sustaining this CO<sub>2</sub> capture process can be efficiently exploited for raising HP steam and producing electricity in a Rankine cycle.

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*Keywords:* Cement; Ca-Looping; CO<sub>2</sub> capture; Post-combustion; Retrofitting

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## 1. Introduction

Cement production is a highly energy-intensive industry and in 2015 was responsible for about 8% of the world's anthropogenic CO<sub>2</sub> emissions [1]. Greenhouse gas emissions for the cement production come from fossil fuel combustion and limestone calcination: about 60% of the CO<sub>2</sub> emissions in a cement kiln comes from the calcination of the limestone contained in the raw material, whereas the remaining 40% is generated by the combustion of fossil fuels needed for sustaining limestone calcination and clinker production.

### Nomenclature

$e_{CO_2}$	Specific CO <sub>2</sub> emissions [ $kg_{CO_2}/t_{clk}$ ]
$P_e$	Net plant absorbed power [ $MW_e$ ]
$q$	Specific fuel consumption [ $MJ_{LHV}/t_{clk}$ ]
<u>Subscripts</u>	
carb	Carbonator
cem	Cement
clk	Clinker
e	Electric
eq	Equivalent
ref	Referent plant (without CO <sub>2</sub> capture system)
th	Thermal
<u>Greek letters</u>	
$\eta$	Efficiency
<u>Acronyms</u>	
ASU	Air separation unit
CaL	Calcium-Looping
CCS	Carbon capture and storage
CFB	Circulating fluidized bed
CPU	CO <sub>2</sub> purification unit
ECRA	European Cement Research Academy
IL	Integration level
LHV	Lower heating value
SPECCA	Specific primary energy for CO <sub>2</sub> avoided [ $MJ_{LHV}/kg_{CO_2}$ ]

Several methods have been proposed to reduce CO<sub>2</sub> emissions in the cement industry: (i) the use of low carbon fossil fuels (e.g. natural gas), (ii) the use of carbon neutral fuels (e.g. biomass), (iii) improvements in thermal efficiency (e.g. increasing the number of preheating stages), (iv) reduction of the clinker/cement ratio by increasing the content of additives in the final cement [2]. However, the CO<sub>2</sub> emission reduction potential of these measures is limited because they do not tackle the issue of the emissions derived from limestone calcination. Therefore, Carbon Capture and Storage (CCS) techniques are necessary to achieve significant cuts in CO<sub>2</sub> emissions in a cement production process [3].

In the cement sector, several CO<sub>2</sub> capture technologies have been investigated so far, including oxy-combustion, post-combustion absorption, membranes and Calcium-Looping [3–7]. This work focuses on the process integration of the Ca-Looping (CaL) process into a cement kiln and summarizes the study presented by De Lena et al in [8]. This capture technology is one of the most promising ones for CO<sub>2</sub> capture in cement kilns [9–12]. The CaL concept was originally proposed by Shimizu et al (1999) [13] and it is based on the reversible reaction between CaO and CO<sub>2</sub> at high temperature. CO<sub>2</sub>-rich gases come into contact with CaO-rich solids in the carbonator (Fig. 1) where the exothermic carbonation reaction occurs (Eq. (1)). Solids containing the CaCO<sub>3</sub> formed by the carbonation reaction are sent to a second reactor (the calciner, Fig. 1) for regeneration at a temperature of 920°C. The high temperatures required to withstand the calcination reaction in a CO<sub>2</sub>-rich environment are guaranteed by the combustion of coal

with an oxygen-rich mixture, so as not to nullify the carbon separation. This CaL process has been successfully demonstrated up to 1-2 MW<sub>th</sub> scale [14,15] in operating conditions representative of the integration in coal-fired power plants, where flue gas has lower CO<sub>2</sub> concentration compared to cement kilns.



One of the inherent advantages of this technology is that most of the fuel chemical energy introduced into the calciner can be recovered as high temperature heat (650°C) in the cooled carbonator and potentially converted into electricity with high efficiency. However, a continuous make-up of fresh limestone is needed for (i) avoiding the build-up of inert species (ash and CaSO<sub>4</sub> generated by coal combustion) and (ii) keeping a proper activity of the sorbent. When the CaL technology is integrated into the cement production process, this disadvantage is compensated by the large availability of on-site limestone, which is first used as sorbent for the CaL process, and then for the production of clinker. For the same reason, cement kilns are often proposed as sinks for the spent CaO from CaL units integrated in power plants, where the CaL purge can be valorized by producing clinker with lower CO<sub>2</sub> emissions and lower fuel consumption without significant modifications to the cement kiln equipment [16].

## 2. Process description

The “Tail-End”, configuration object of this work is presented in Fig. 1. The CaL system is integrated downstream of the clinker production process, to capture the CO<sub>2</sub> generated in the clinker burning process [6,8,12,17–19]. This CaL application has been recently demonstrated in two different experimental facilities, at 30 kW<sub>th</sub> and 200 kW<sub>th</sub> scale [20,21].

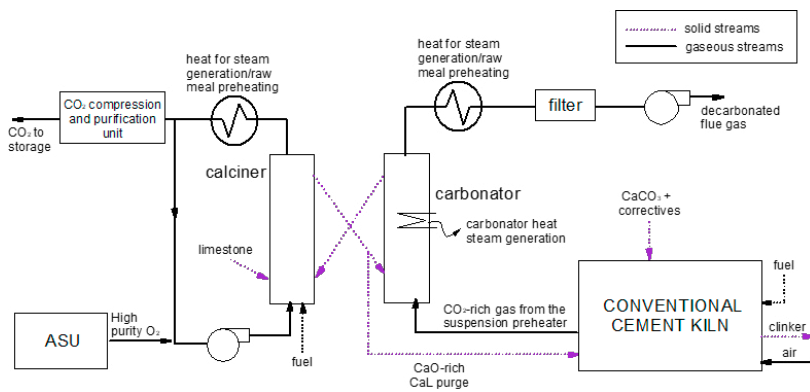


Fig. 1. Integration of the Tail-end CaL process in a cement kiln

The clinker production process is designed on the Best Available Technique (BAT) standard as defined in the European BREF document for the manufacture of cement [22]. This process constitutes the core of the reference cement kiln without CO<sub>2</sub> capture, and it remains practically unchanged when integrated with the CaL process. The cement kiln considered in this work has a production capacity of around 3000 t/d (which is a representative size for a European cement kiln) and it is made up of (i) clinker cooler, (ii) rotary kiln, (iii) pre-calciner, (iv) five-stage suspension preheater. The main process assumptions used for carrying out the simulation are reported in Tab. 1 and have been defined according to the model developed by the German Cement Works Association VDZ for the European Cement Research Academy (ECRA) [23] and the IEA Greenhouse Gas R&D Programme [3]. More details on the process model are available in the public document [24].

Raw meal used for clinker production consists mainly of CaCO<sub>3</sub> and oxides of Si, Fe e Al, which are introduced through additional raw materials in order to achieve the targeted composition given in Tab. 2. The raw material is fed into the top of the preheater and preheated by direct contact with hot gases from the pre-calciner.

The preheated solids are fed to the pre-calciner, where about 94% of the CaCO<sub>3</sub> contained in the raw meal is converted into CaO and CO<sub>2</sub>. Energy needed for this endothermic reaction is provided by the combustion of coal (Tab.

2). The almost totally calcined solids coming out of the pre-calciner at about 860°C, enter the rotary kiln. In the rotary kiln the CaCO<sub>3</sub> calcination is completed and, as the solids moves towards the rotary kiln burner, the different clinker phases are formed thanks to the high temperature. The hot clinker reaches a temperature of about 1450°C at the end of the kiln before being discharged to the clinker cooler, where the clinker is rapidly cooled down to minimize the formation of undesired crystalline phases.

Tab. 1. Main assumptions used for the process simulations

Clinker production process		CaL System	
Ambient temperature [°C]	15	Carbonator outlet temperature [°C]	650
Clinker production [tpd]	2825	Carbonator solid inventory [kg/m <sup>2</sup> ]	1000
Clinker/cement ratio [-]	0.737	Gas superficial velocity at carbonator inlet [m/s]	5
Electric consumption of auxiliaries [kWh/t <sub>cem</sub> ]	97	Δp gas in the carbonator [kPa]	20
Coal input to pre-calciner [MJ <sub>LHV</sub> /kg <sub>clik</sub> ]	0.84	Calciner outlet temperature [°C]	920
Coal input to rotary kiln [MJ <sub>LHV</sub> /kg <sub>clik</sub> ]	1.21	Recycle gas temperature [°C]	400
Pre-calciner outlet temperature [°C]	861.8	Oxygen concentration in oxidant flow [%vol.]	50
Rotary kiln gas outlet temperature [°C]	1078.5	Oxygen preheating temperature [°C]	150
Clinker temperature at rotary kiln outlet [°C]	1457.0	Oxygen concentration in calciner off-gas [%vol.]	5
Clinker temperature at clinker cooler outlet [°C]	114.9	<b>ASU</b>	
Calcination degree at pre-calciner outlet [%]	94.2	Oxygen purity [%vol.]	95
<b>Steam cycle</b>		Electric consumption [kWh/t <sub>CO2</sub> ]	226
Steam pressure at turbine inlet [bar]	100	<b>CPU</b>	
Steam temperature at turbine inlet [°C]	530	CO <sub>2</sub> purity [%vol.]	>95%
Condensing pressure [bar]	0.07	Electric consumption [kWh/t <sub>CO2</sub> ]	112
Steam turbine isentropic/ mechanical-electric efficiency [%]	85.7 / 97	CO <sub>2</sub> delivery pressure [bar]	150

Tab. 2. Composition of coal and raw meal

Coal composition [%wt.]		Raw meal composition [%wt.]	
C	69.00	CaCO <sub>3</sub>	79.08
H	4.00	SiO <sub>2</sub>	13.77
S	0.50	Al <sub>2</sub> O <sub>3</sub>	3.33
N	0.48	Fe <sub>2</sub> O <sub>3</sub>	2.01
O	9.00	MgCO <sub>3</sub>	1.53
Moisture	0.50	Moisture	0.28
Ash	16.50		
Lower Heating Value [MJ/kg]	27.15		

Exhaust gas exiting the preheater at about 330°C is fed to the CFB carbonator of the CaL system, where 90% of the CO<sub>2</sub> is captured by the exothermic carbonation reaction between CaO and CO<sub>2</sub>. The carbonator is a cooled reactor operating at a temperature of 650°C [16]. The CaCO<sub>3</sub>-rich solids exiting the carbonator are sent to the calciner, where calcination at a temperature of 920°C is sustained by oxy-combustion of coal [25]. A cryogenic Air Separation Unit (ASU) produces 95% pure oxygen to be used as oxidant in the CaL calciner. The CO<sub>2</sub>-rich stream, generated in the calciner of the CaL process, is cooled down and sent to a CO<sub>2</sub> purification and compression unit (CPU), which allows achieving a CO<sub>2</sub> purity of >95% at a pressure of 110 bar. The main assumptions adopted for these two components are reported in Tab. 1.

The decarbonized flue gas stream exiting the carbonator is cooled down and sent to the raw mill section for drying coal and raw meal.

The CaL purge is also cooled down and sent to the raw mill, where it is mixed and grounded with the raw meal. This operation is necessary because the particles coming from the CaL system, have a size of about 200  $\mu\text{m}$ , much larger than that typically used in cement kilns (about 30  $\mu\text{m}$ ). The *Integration Level* (IL) is defined as the percentage of Ca entering the clinker burning process with the CaL purge with respect to the total Ca fed to the plant.

Given both the amount and the temperature level of the waste heat available in the CaL system, the most efficient and cost-effective technology for electricity production is a Heat Recovery Steam Cycle. In this way, it is possible to generate electricity to partly compensate the electric consumption of the cement kiln and CO<sub>2</sub> capture section auxiliaries (ASU, CPU, fans, etc.).

### 3. Methodology and key performance indicators

Heat and mass balances have been calculated with the Polimi in-house code GS (Gas-Steam cycle) [26]. The CO<sub>2</sub> capture efficiency in the carbonator is calculated with the model developed by Romano (2012) [27].

The performances of the cement kiln with and without CO<sub>2</sub> capture system are evaluated by the following key performance indicators:

- *Direct fuel consumption* ( $q$ ) [ $\text{MJ}_{\text{LHV}}/\text{t}_{\text{clik}}$ ]: the primary energy consumed through coal combustion in the cement kiln per unit of clinker produced.
- *Indirect fuel consumption* ( $q_e$ ) [ $\text{MJ}_{\text{LHV}}/\text{t}_{\text{clik}}$ ]: the primary energy consumption associated to the net electric consumption in the cement kiln ( $P_e$ ). This index depends on the reference electrical efficiency ( $\eta_{\text{ref},e}$ ), which depends on the technology and fuel used for the power generation (Eq. (2))

$$q_e = \frac{P_e}{\eta_{\text{ref},e}} \quad (2)$$

- *Equivalent fuel consumption* ( $q_{eq}$ ) [ $\text{MJ}_{\text{LHV}}/\text{t}_{\text{clik}}$ ]: the sum of the direct ( $q$ ) and indirect ( $q_e$ ) fuel consumptions.
- *Direct CO<sub>2</sub> emissions* ( $e_{\text{CO}_2}$ ) [ $\text{kg}_{\text{CO}_2}/\text{t}_{\text{clik}}$ ]: the amount of CO<sub>2</sub> directly emitted at the stack of the cement kiln per unit of clinker produced.
- *Indirect CO<sub>2</sub> emissions* ( $e_{\text{CO}_2,e}$ ) [ $\text{kg}_{\text{CO}_2}/\text{t}_{\text{clik}}$ ]: CO<sub>2</sub> emissions associated to the production of the electricity consumed in the full process ( $P_e$ ) per unit of clinker produced (Eq. (3)).

$$e_{\text{CO}_2,e} = P_e \cdot e_{\text{ref},e} \quad (3)$$

where the term  $e_{\text{ref},e}$  refers to the specific emissions of the reference power generation scenario considered.

- *Equivalent CO<sub>2</sub> emissions* ( $e_{\text{CO}_2,eq}$ ) [ $\text{kg}_{\text{CO}_2}/\text{t}_{\text{clik}}$ ]: the sum of the direct ( $e_{\text{CO}_2}$ ) and the indirect ( $e_{\text{CO}_2,e}$ ) emissions.
- *Specific Primary Energy Consumption for CO<sub>2</sub> Avoided (SPECCA, Eq. (4))* [ $\text{MJ}_{\text{LHV}}/\text{kg}_{\text{CO}_2}$ ], indicates the additional equivalent primary energy consumption to avoid the emission of a unit mass of equivalent CO<sub>2</sub> with respect to the reference cement kiln without CO<sub>2</sub> capture system.

$$\text{SPECCA} = \frac{q_{eq} - q_{eq,ref}}{e_{\text{CO}_2,eq,ref} - e_{\text{CO}_2,eq}} \quad (4)$$

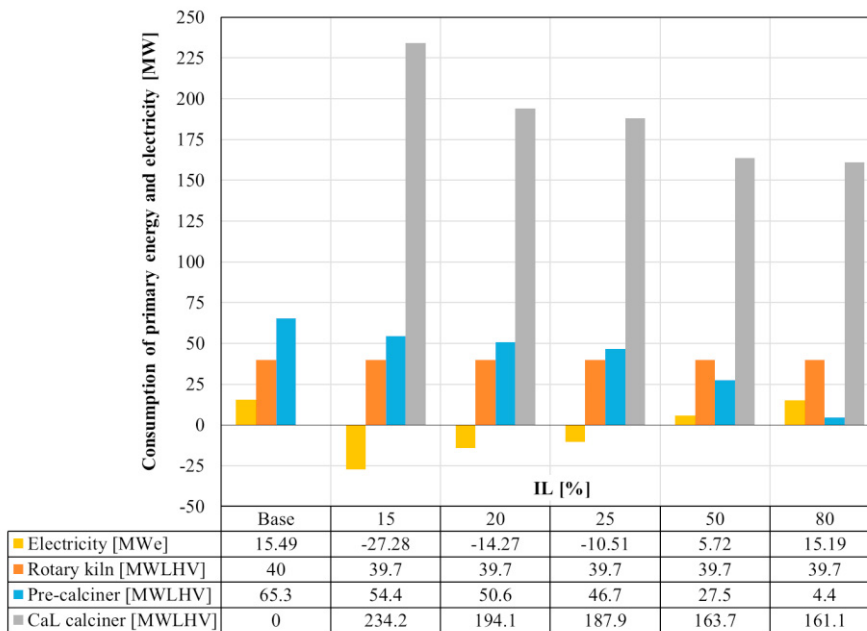
### 4. Results

Details of the results for the case with IL 50%, inventory of solids in the carbonator of 1000  $\text{kg}/\text{m}^2$  and efficiency of the carbonator 90% are given in Tab. 3, which also shows the results for the reference case without CCS.

The total fuel thermal input increases by about 120% compared to the reference case. Fuel consumption in the rotary kiln remains roughly constant, whereas the pre-calciner fuel consumption is reduced by about 58% with respect to the reference cement kiln, because of the replacement of CaCO<sub>3</sub> in the raw meal with CaO from the CaL solid purge. The fuel consumption in the calciner of the CaL process accounts for 30% of the total fuel consumption of the plant. This large increase in the total fuel consumption is due to the fact that the CO<sub>2</sub> from the pre-calciner undergoes a second calcination in the calciner of the CaL system, after the first calcination in the pre-calciner of the cement kiln. Fig. 2 shows the trends of primary energy consumption and electrical consumption (negative if there is a net production of electricity) for different integration levels.

Tab. 3. Main results of the simulation for the case with IL 50%

	Cement kiln without CCS		Case IL50-W1000	
Total heat input (q), $MW_{LHV} - MJ_{LHV}/kg_{clk}$	105.4	3.22	230.9	7.06
Heat input in rotary kiln, $MW_{LHV} - MJ_{LHV}/kg_{clk}$	40.0	1.22	39.7	1.21
Heat input in pre-calciner, $MW_{LHV} - MJ_{LHV}/kg_{clk}$	65.3	2.00	27.5	0.84
Heat input in CaL calciner, $MW_{LHV} - MJ_{LHV}/kg_{clk}$	-	-	163.7	5.01
<i>Power balance</i>				
Gross steam turbine electricity production, $MW_e - kWh_e/t_{clk}$	-	-	41.58	353.2
Net electricity consumption, $MW_e - kWh_e/t_{clk}$	15.49	131.6	5.72	48.6
Net electricity consumption, $kWh_e/t_{cem}$		97.0		35.8

Fig. 2. Electricity consumption and direct fuel consumption of cement kiln without CCS and with CO<sub>2</sub> capture by CaL process with different values of IL

For the cases with IL between 15 and 25%, there is a net power production (respectively 65 e 170 kWh<sub>e</sub>/t<sub>cem</sub>) exported to the grid. In contrast, for cases with IL of 50 and 80% a net electricity consumption of 36 e 95 kWh<sub>e</sub>/t<sub>cem</sub> respectively is obtained. These values are comparable to the power consumption of the cement kiln without CCS (97 kWh<sub>e</sub>/t<sub>cem</sub>).

Finally the key performance indicators defined in Section 3 are reported in Tab. 4 for the reference case and for the CaL case with IL 50%. The analysis includes two different scenarios for the power production: (i) a state of the art pulverized coal ultra-supercritical power plant and (ii) the European energy mix [28].

Direct CO<sub>2</sub> emissions are reduced by 91.2% with respect to the cement kiln without CCS. For the case selected, the Tab. 4 shows positive indirect CO<sub>2</sub> emissions, associated with a net power consumption. This results in a reduction of equivalent CO<sub>2</sub> emissions between 88.3% and 90.1%, which is lower than direct emission reduction.

SPECCA between 3.70 and 3.93 MJ/kg<sub>CO2</sub> have been obtained.

Tab. 4. Key performance indicators for the case with IL 50% under two different reference scenarios for electricity production

	Cement kiln without CCS		Case IL 50%-Ws 1000	
Direct fuel consumption (q), $MW_{LHV} - MJ_{LHV}/kg_{clik}$	105.4	3.22	230.9	7.06
Direct CO <sub>2</sub> emission ( $e_{CO_2}$ ), $kg/s - kg_{CO_2}/t_{clik}$	28.3	865.2	2.5	76.3
Direct emission reduction, %	-	-	91.2%	
<i>State of the art pulverized coal ultra-supercritical power plant (<math>\eta_e = 44.2\%</math>, <math>e_{CO_2} = 770 \text{ kg}_{CO_2}/MWh_e</math>)</i>				
Indirect fuel consumption ( $q_e$ ), $MW_{LHV} - MJ_{LHV}/kg_{clik}$	35.0	8.98	12.9	0.39
Equivalent fuel consumption ( $q_{eq}$ ), $MW_{LHV} - MJ_{LHV}/kg_{clik}$	140.4	4.30	243.8	7.45
Indirect CO <sub>2</sub> emission ( $e_{CO_2,e}$ ), $kg/s - kg_{CO_2}/t_{clik}$	3.3	101.3	1.2	37.2
Equivalent CO <sub>2</sub> emission ( $e_{CO_2,eq}$ ), $kg/s - kg_{CO_2}/t_{clik}$	31.6	966.6	3.7	113.5
Equivalent emission reduction, %	-	-	88.3%	
SPECCA, $MJ_{LHV}/kg_{CO_2}$	-	-	3.70	
<i>Energy mix (2015) EU-28 non-CHP (<math>\eta_e = 45.9\%</math>, <math>e_{CO_2} = 262 \text{ kg}_{CO_2}/MWh_e</math>)</i>				
Indirect fuel consumption ( $q_e$ ), $MW_{LHV} - MJ_{LHV}/kg_{clik}$	33.7	8.65	12.3	0.38
Equivalent fuel consumption ( $q_{eq}$ ), $MW_{LHV} - MJ_{LHV}/kg_{clik}$	139.1	4.26	243.2	7.44
Indirect CO <sub>2</sub> emission ( $e_{CO_2,e}$ ), $kg/s - kg_{CO_2}/t_{clik}$	1.1	34.5	0.4	12.6
Equivalent CO <sub>2</sub> emission ( $e_{CO_2,eq}$ ), $kg/s - kg_{CO_2}/t_{clik}$	29.4	899.7	2.9	88.9
Equivalent emission reduction, %	-	-	90.1%	
SPECCA, $MJ_{LHV}/kg_{CO_2}$	-	-	3.93	

## 5. Conclusions

In this work, the integration of a tail-end Calcium looping (CaL) process into a cement kiln is assessed, where the CaL carbonator is used as an end-of-pipe unit to capture the CO<sub>2</sub> produced in the clinker production process. The results obtained have shown that the system provides direct CO<sub>2</sub> emission reductions at the cement kiln of more than 90%. However, this process requires intrinsically high fuel consumptions, due to the double calcination needed for the CO<sub>2</sub> originating from limestone decomposition. The heat recovered by the CaL system makes it possible to produce electricity, which in case with integration level of 50% offsets the power consumption of the ASU and CPU. The results show that SPECCA is between 3.70 e 3.93  $MJ_{LHV}/kg_{CO_2}$ . More details on balances, scenarios and retrofitting possibilities can be found in [8].

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