

# **Calcium looping combustion for high-efficiency low-emission power generation**

*Dawid P. Hanak\*, Vasilije Manovic*

*Combustion and CCS Centre, Cranfield University,*

*Bedford, Bedfordshire, MK43 0AL, UK*

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*Corresponding author: \*Dawid P. Hanak, [d.p.hanak@cranfield.ac.uk](mailto:d.p.hanak@cranfield.ac.uk)*

## ABSTRACT

High-temperature solid looping technologies, such as calcium looping and chemical looping combustion are regarded as emerging CO<sub>2</sub> capture technologies with potential to reduce the net efficiency penalties associated with CO<sub>2</sub> separation. Importantly, high-temperature operation of these technologies allows utilisation of the high-grade heat for power generation. Building on these emerging technologies, this study intended to establish a new class of high-temperature solid looping combustion technologies for high-efficiency low-emission power generation called calcium looping combustion. Such combustion technology comprises a combustor, as a primary source of heat for indirect heating in a calciner, and a carbonator where CO<sub>2</sub> is separated from flue gas leaving the combustor; hence high-grade heat, which can be used for power generation, and a concentrated CO<sub>2</sub> stream, which can be either utilised or permanently stored, are generated. The techno-economic performance of calcium looping combustion was comparable to a conventional coal-fired power plant. Depending on whether the concentrated CO<sub>2</sub> stream is utilised elsewhere or permanently stored, calcium looping combustion was characterised with a net efficiency gain of 0.7%<sub>HHV</sub> points or a net efficiency penalty of 2.4%<sub>HHV</sub>, respectively. Additionally, the cost of CO<sub>2</sub> avoided for calcium looping combustion was estimated to be 10.0 €/tCO<sub>2</sub> and 33.9 €/tCO<sub>2</sub>, respectively. Therefore, similarly to chemical looping combustion, calcium looping combustion introduced in this study is a viable high-efficiency low-emission power generation technology that produces a concentrated CO<sub>2</sub> stream with no efficiency penalty associated with CO<sub>2</sub> separation.

**Key Words:** Novel power generation system; coal-fired power plant; process modelling and simulation; techno-economic analysis; calcium looping; decarbonisation

# 1 INTRODUCTION

To significantly reduce the risks and impacts of climate change, the global mean temperature increase needs to be held well below 2°C and efforts to limit it to 1.5°C above the pre-industrial levels need to be pursued [1]. Carbon capture and storage (CCS) has been recognised to be essential for decarbonisation of the power sector, which is a critical step towards reduction of global greenhouse gas emissions [2,3], as it can enable a low-emission and flexible power generating capacity [4,5]. Yet, regardless of the recent progress in both oxy-combustion and chemical solvent scrubbing technologies [6,7], these have been predicted to reduce the net thermal efficiency of the conventional coal-fired power plant (CFPP) by 7–13% points [8–10]. This corresponds to an increase in the cost of electricity of at least 60% [11–13]. Calcium looping (CaL), which is based on the reversible carbonation reaction of lime with CO<sub>2</sub>, is regarded as an emerging CO<sub>2</sub> capture technology and has already been demonstrated at a scale of up to 1.9 MW<sub>th</sub> [8,14]. In the state-of-the-art configuration of CaL, the heat required for sorbent regeneration is provided via oxy-combustion of fuel directly in the calciner to ensure high purity of the concentrated CO<sub>2</sub> stream. Retrofits of such a high-temperature solid looping technology to CFPPs were predicted to impose a net efficiency penalty of 5–8% points [8,14], which is considerably lower compared to the figures reported for the mature CO<sub>2</sub> capture technologies. The main source of the parasitic load in CaL is the power required to drive the compressors in both the CO<sub>2</sub> compression unit (CCU) and the air separation unit (ASU) [15,16]. The requirement for the former unit, and hence its contribution to the net efficiency penalty, depend on whether the concentrated CO<sub>2</sub> stream is permanently stored or utilised, for example, for chemicals or fuels production [17,18]. For this reason, the main reduction in the efficiency penalty

associated with CaL can be primarily achieved via utilising alternative options to provide heat for sorbent regeneration and thus by avoiding the need for oxygen production in the energy-intensive ASU.

The main alternative heat sources to drive the calcination process include chemical looping [19–22], which uses oxygen carriers to transfer oxygen from air to the fuel, and indirect heat transfer from a combustor via solid heat carriers [15,23], heat transfer wall [15,24] or heat pipes [25,26]. The combined calcium and chemical looping system includes an additional reactor – an air reactor – in which the oxygen carrier reacts with oxygen in the air, forming a metal oxide. The oxygen carrier is then reduced by the gaseous fuel in a fuel reactor, providing heat for calcination. This system can operate as either a single- or dual-loop process [20,22], depending on whether the oxygen carrier is mixed with the sorbent and reduced directly in the calciner, or is handled separately in another loop. In the latter case, the heat is transferred to the calciner indirectly. The efficiency penalty associated with the combined calcium and chemical looping process was estimated to fall between 3.6–6.9% points and 5.2–6.3% points for the single- and dual-loop process [20,21], respectively. Although the net efficiency of such a process was found to be 2.5–3% points higher than that of CaL retrofitted to the same reference CFPP, the required solids looping rate to achieve the same CO<sub>2</sub> capture level was at least one order of magnitude higher than that in CaL [20]. This may impose a further efficiency penalty associated with the requirement for handling more solids. An alternative to the combined calcium and chemical looping process is the concept of calcination driven by heat carriers [15,23]. This process involves an additional circulating fluidised bed combustor in which fuel is burned in an excess amount of air. The bed material, which is a dense solid material such as aluminium (III) oxide or deteriorated sorbent,

is heated up directly in the combustor, separated from the flue gas stream, and then fed to the calciner to provide heat for sorbent regeneration. However, it is still not clear whether continuous separation of the sorbent and the heat carrier based on the difference in their densities would be possible at the required scale. Nevertheless, the net efficiency of the retrofit scenario based on such a system is claimed to be 2.2% points higher than that of the state-of-the-art CaL retrofit [15]. In order to avoid the need for solids segregation, heat for the sorbent regeneration can be supplied from an external source, such as an additional circulating fluidised bed combustor, via either a heat transfer wall [15,24] or heat pipes [25,26]. Recently, the latter option has been experimentally proven [25–27]. Utilisation of an indirectly-heated calciner was reported to have 1.6% points higher net efficiency compared to a CaL retrofit [15], and to result in a net efficiency penalty of 1.5–3.5% points, if CO<sub>2</sub> compression was not accounted for [26]. Hence, this option for providing heat to drive the calcination process appears to be the most appealing at the moment.

Due to high-temperature operation of CaL and, thus availability of a large amount of high-grade heat that can be utilised for power generation in the secondary steam cycle, its retrofits to CFPPs were shown to increase the net power output by around 50–80% compared to the net power output of the reference CFPP without CO<sub>2</sub> capture [16,28–31]. Therefore, in the retrofit scenarios, CaL can be seen as a secondary power boiler in which part of the heat input from the oxy-combustion of fuel, or an external heat source, is used for sorbent regeneration in the calciner and is then recovered in the carbonator. Similarly to chemical looping combustion, which is also regarded as an emerging CO<sub>2</sub> separation technology [32], CaL with an indirectly-heated calciner and the combustor can be treated as the primary heat sources for power generation and, hence can be seen as a novel high-temperature

solid looping combustion technology. Therefore, by using such combustion technology for high-pressure steam generation, the opportunity arises to develop novel high-efficiency low-emission power generation technologies that will be able to meet the specific emission target of 100 g/kW<sub>el</sub>h beyond 2050 and ensure security of future electricity supply at affordable prices. The current literature [15,25,26], however, provides scarce information on the thermodynamic and no information on the economic feasibility assessment of such novel power generation technologies. Moreover, the available configurations assume that the sorbent in the indirectly-heated calciner is fluidised either by CO<sub>2</sub> released during calcination (self-fluidisation) [25,26] or by steam [15]. Although self-fluidisation was found to be feasible [33], use of the external fluidisation medium improves fluidisation and mixing of sorbent particles and, hence heat and mass transfer within the calciner. Furthermore, the use of steam as fluidising medium would reduce the calcination temperature, increasing the heat transfer rate between the calciner and the combustor. Yet, application of steam would increase the calciner heat requirement and, if drawn from the steam cycle, would reduce the gross power output, affecting the efficiency of the entire process [33]. In addition, to maintain the desired calcination temperature, the need for solid-solid heat exchange was indicated [25,26].

This study intends to establish a new class of high-temperature solid looping combustion technologies for high-efficiency low-emission power generation by evaluating the feasibility of a calcium looping combustion (CaLC) process that, while producing a concentrated CO<sub>2</sub> stream, has a higher net thermal efficiency than, and comparable cost of electricity to, conventional CFPPs without CO<sub>2</sub> capture. A techno-economic analysis and parametric studies on the key design parameters are

performed to assess the process feasibility. The techno-economic performance of CaLC is benchmarked against the conventional CFPP in the scenarios considering utilisation (without CCU) and permanent storage (with CCU) of the concentrated CO<sub>2</sub> stream.

## **2 PROCESS DESCRIPTION**

### **2.1 Conventional coal-fired power plant**

The conventional 580 MW<sub>el</sub> CFPP is used as a reference scenario in this study [34–36]. This CFPP comprises a power boiler, which operates with the equivalence ratio of 1.1, with NO<sub>x</sub> and SO<sub>x</sub> emission control equipment and the electrostatic precipitator. Heat from coal combustion in the power boiler is utilised to raise the steam for the reheated regenerative steam cycle operating at the supercritical steam conditions. The steam generator consists of the heat exchange sections, which are the primary, secondary, and reheat superheaters, as well as the economiser. Both the live (242.3 bar) and reheat steam generated in these sections are leaving the boiler at the temperature of 593.3°C. The steam generated in the power boiler is fed to the steam turbine section that comprises high- (HP), intermediate- (IP) and low-pressure (LP) extraction condensing steam turbines. Moreover, part of the steam from the turbine sections is drawn to feed the main feedwater heating train. This consists of five LP feedwater heaters, the last one of which is called deaerator and is a mixed feedwater heater, and three HP feedwater heaters. Such conventional CFPP was found to deliver the net power output of 552.7 MW<sub>el</sub> at the net thermal efficiency of 38.0%<sub>HHV</sub>.

### **2.2 Calcium looping combustion**

A core of the CaLC process (Figure 1) is the CaL process that was proposed by Shimizu et al. [37] as a post-combustion CO<sub>2</sub> capture system for fossil fuel power

plants. The CaLC process comprises two interconnected fluidised bed reactors – a carbonator and a calciner – and a circulating fluidised bed combustor, all operating at atmospheric pressure. As opposed to the state-of-the-art CaL configuration, in which the heat for sorbent regeneration is provided via oxy-combustion of fuel [8,14], the CaLC process utilises indirect heat transfer from the combustor to the calciner that can be facilitated via either a heat transfer wall [15] or heat pipes [25,26]. Under the initial design basis (Table 1), the operating temperature of the combustor is maintained by burning coal in an excess amount of air to ensure near-complete combustion. To maximise combustion efficiency, the combustion air is preheated by the concentrated CO<sub>2</sub> stream and the hot sorbent stream leaving the calciner cyclone. Having been cooled prior to being fed to the CCU, a fraction of the cold concentrated CO<sub>2</sub> stream is diverted to the fluidising fan and then preheated by the hot concentrated CO<sub>2</sub> stream leaving the calciner cyclone to minimise the calciner heat requirement. To further reduce the amount of heat required to be transferred from the combustor to the calciner, heat carried by the purge stream is recovered by the cooling air and is then used to preheat the fresh limestone make-up stream. The flue gas leaving the combustor is then cooled and, without being desulphurised, is fed directly to the carbonator for CO<sub>2</sub> and SO<sub>x</sub> removal. Importantly, due to high affinity of the sorbent to SO<sub>x</sub> and the high Ca:S ratios in the system, the SO<sub>x</sub> capture level is nearly 100% [38–41].

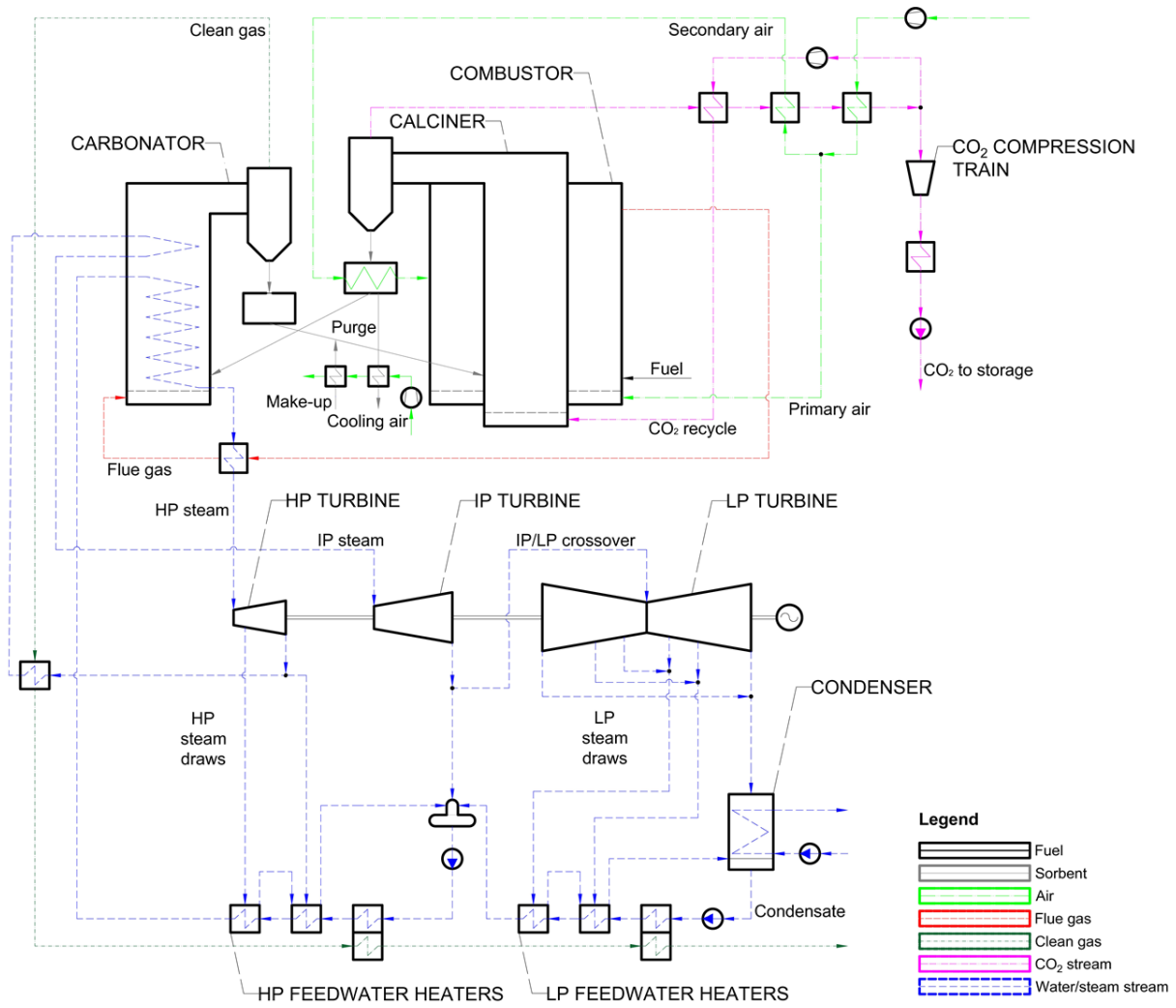


**Figure 1: Schematic of calcium looping combustion with CO<sub>2</sub> compression unit**

The high-grade heat available in the carbonator, as well as in the clean gas and flue gas streams, is utilised to raise live steam for, and for feedwater heating in, the supercritical steam cycle. The live steam enters the HP turbine cylinder at 593.3°C and 242.3 bar, and is expanded to 49.0 bar. It is reheated by the clean gas stream and in the carbonator, before it is sent to the IP turbine cylinder, and subsequently to two LP turbine cylinders. To enhance the thermal efficiency of the steam cycle, steam extracted from the turbine cylinders and the remaining heat carried by the clean gas are used for feedwater heating. The feedwater heating train in the CaLC process comprises four LP feedwater heaters, the last one of which is a mixed feedwater heater (deaerator), and three HP feedwater heaters. The characteristics of

this steam cycle are comparable to those in the conventional 580 MW<sub>el</sub> CFPP that is used as a reference power generation system in this study.

**Table 1: Initial design conditions and process model assumptions for calcium looping combustion**

| Parameter                              | Value                                                                                                                                                                      |
|----------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| <b>Carbonator</b>                      | Stoichiometric reactor. Average conversion model by Rodríguez et al. [42] with deactivation curve from Sánchez-Biezma et al. [38].                                         |
|                                        | Temperature (°C)                                                                                                                                                           |
|                                        | Pressure drop (mbar)                                                                                                                                                       |
|                                        | Carbonation extent (-)                                                                                                                                                     |
| <b>Calciner</b>                        | Gibbs reactor. Gibbs free energy minimisation model.                                                                                                                       |
|                                        | Temperature (°C)                                                                                                                                                           |
|                                        | Pressure drop (mbar)                                                                                                                                                       |
|                                        | Calcination extent (-)                                                                                                                                                     |
|                                        | Relative make up (Fresh limestone/Sorbent circulation rate) (-)                                                                                                            |
|                                        | Fraction of CO <sub>2</sub> recycled to calciner (-)                                                                                                                       |
| <b>Combustor</b>                       | Gibbs reactor. Gibbs free energy minimisation model. Coal composition adapted from the revised NETL report [34] and its heating value is estimated using Dulong's formula. |
|                                        | Temperature (°C)                                                                                                                                                           |
|                                        | Pressure drop (mbar)                                                                                                                                                       |
|                                        | Equivalence ratio (-)                                                                                                                                                      |
| <b>Supercritical steam cycle</b>       | Design live/reheat steam temperature (°C)                                                                                                                                  |
|                                        | Design live/reheat steam pressure (bar)                                                                                                                                    |
|                                        | Final feedwater temperature (°C)                                                                                                                                           |
|                                        | Feedwater heater terminal temperature difference (°C)                                                                                                                      |
|                                        | Feedwater heater minimum temperature approach (°C)                                                                                                                         |
|                                        | Isentropic efficiency of compressors (%)                                                                                                                                   |
|                                        | Isentropic efficiency of high-pressure steam turbine (%)                                                                                                                   |
|                                        | Isentropic efficiency of intermediate-pressure steam turbine (%)                                                                                                           |
|                                        | Isentropic efficiency of low-pressure steam turbine (%)                                                                                                                    |
|                                        | Isentropic efficiency of pumps (%)                                                                                                                                         |
|                                        | Electrical efficiency of generator (%)                                                                                                                                     |
|                                        | Mechanical efficiency of steam turbines (%)                                                                                                                                |
|                                        | Mechanical efficiency of compressors (%)                                                                                                                                   |
|                                        | Mechanical efficiency of compressors (%)                                                                                                                                   |
| <b>Heat exchanger network</b>          | Sorbent cooler and heater minimum temperature approach (°C)                                                                                                                |
|                                        | Air preheater minimum temperature approach (°C)                                                                                                                            |
|                                        | CO <sub>2</sub> preheater minimum temperature approach (°C)                                                                                                                |
|                                        | CO <sub>2</sub> preheater minimum temperature approach (°C)                                                                                                                |
| <b>CO<sub>2</sub> compression unit</b> | Polytropic efficiency of CO <sub>2</sub> compressors (%)                                                                                                                   |
|                                        | Isentropic efficiency of CO <sub>2</sub> pump (%)                                                                                                                          |
|                                        | Mechanical efficiency of compressors and pump (%)                                                                                                                          |
|                                        | Intercooling temperature (°C)                                                                                                                                              |
|                                        | CO <sub>2</sub> delivery pressure (bar)                                                                                                                                    |

### 3 PROOF OF CONCEPT

#### 3.1 Considerations

The process model of the proposed CaLC process has been developed by utilising the existing process models of CaL and supercritical steam cycle in the conventional 580 MW<sub>el</sub> CFPP that have been previously developed in Aspen Plus® and successfully benchmarked against data available in the literature [16,35,36]. The initial design conditions and process model assumptions are summarised in Table 1. The techno-economic performance of the CaLC process is evaluated in two scenarios with concentrated CO<sub>2</sub> sent for utilisation (without CCU) or permanent storage (with CCU), and is benchmarked against the performance of the conventional 580 MW<sub>el</sub> CFPP. A parametric study is then conducted to evaluate the effect of design conditions on the techno-economic performance of the CaLC process.

The thermodynamic performance of the CaLC process is characterised using the key performance indicators that are commonly used to assess the performance of conventional CFPPs. These primarily are the net power output ( $\dot{W}_{net}$ ) and net thermal efficiency ( $\eta_{th}$ ), which is defined in Eq. (1) as the ratio of the net power output and the heat input from fuel combustion ( $\dot{Q}_{fuel}$ ). In addition, the net efficiency penalty ( $EP$ ) defined in Eq. (2) is calculated to benchmark CaLC performance against the reference CFPP. As CaLC can be seen as a novel power boiler, the boiler thermal efficiency ( $\eta_b$ ), which is defined in Eq. (3) as the ratio of the heat transferred to the working medium in the steam cycle ( $\dot{Q}_{sc}$ ) and the heat input from fuel combustion, is also estimated. The numerator in Eq. (3) is quantified as the amount of heat transferred from the flue gas and clean gas streams, as well as the carbonator to the feedwater and steam in the steam cycle (Figure 1). This definition is also valid for the

reference CFPP, with the difference that the heat is transferred only from the flue gas stream to the feedwater and steam in the steam cycle. Finally, environmental performance is represented in Eq. (4) as the specific CO<sub>2</sub> emissions ( $e_{CO_2}$ ) defined as the ratio of CO<sub>2</sub> emission rate ( $\dot{m}_{CO_2}$ ) and the net power output.

$$\eta_{th} = \frac{\dot{W}_{net}}{\dot{Q}_{fuel}} \quad (1)$$

$$EP = \eta_{th,ref} - \eta_{th,CaLC} \quad (2)$$

$$\eta_b = \frac{\dot{Q}_{sc}}{\dot{Q}_{fuel}} \quad (3)$$

$$e_{CO_2} = \frac{\dot{m}_{CO_2}}{\dot{W}_{net}} \quad (4)$$

The economic performance of the CaLC process is compared with the reference CFPP without CO<sub>2</sub> capture in terms of the levelised cost of electricity ( $LCOE$ ) and the cost of CO<sub>2</sub> avoided ( $AC$ ) that are calculated according to Eq. (5) and Eq. (6) [43–45], respectively.

$$LCOE = \frac{TCR \times FCF + FOM}{\dot{W}_{net} \times CF \times 8760} + VOM + \frac{SFC}{\eta_{th}} \quad (5)$$

$$AC = \frac{LCOE_{capture} - LCOE_{ref}}{e_{CO_2,ref} - e_{CO_2,capture}} \quad (6)$$

These parameters correlate thermodynamic performance indicators, such as net power output, net thermal efficiency, capacity factor ( $CF$ ) and specific CO<sub>2</sub> emissions, with economic performance indicators, such as total capital requirement ( $TCR$ ), variable ( $VOM$ ) and fixed ( $FOM$ ) operating and maintenance costs, specific fuel cost ( $SFC$ ), and the fixed charge factor ( $FCF$ ), which considers the system's lifetime and project interest rate.

The capital cost of the reference CFPP is determined using the exponential method function [46] with economic data presented in Table 2. Taking the capital cost for air-

fired circulating moving-bed with regenerative carbonate process system as the reference capital cost ( $C_0$ ) [47], Eq. (7), which is a modification of the correlation developed by Romano et al. [48], is employed to estimate the total capital cost of the CaLC process ( $C$ ), considering the volume of the reactors ( $V$ ) and the heat input to the combustor ( $\dot{Q}_{comb}$ ) with scaling factors of 0.67 and 0.9, respectively. Moreover, the parameter representing the fraction of the total cost of the combustor associated with the heat transfer surfaces ( $\alpha$ ) is assumed to be 0.85.

$$C = C_0 \left[ \alpha \left( \frac{\dot{Q}_{comb}}{\dot{Q}_{0,comb}} \right)^{SF,Q} + (1 - \alpha) \left( \frac{V_{comb}}{V_{0,comb}} \right)^{SF,V} + (1 - \alpha) \left( \frac{V_{carb}}{V_{0,carb}} \right)^{SF,V} + (1 - \alpha) \left( \frac{V_{calc}}{V_{0,calc}} \right)^{SF,V} \right] \quad (7)$$

**Table 2: Economic model assumptions**

| Parameter                          |                                                                               | Value  |
|------------------------------------|-------------------------------------------------------------------------------|--------|
| Reference coal-fired power plant   | Reference equipment capital cost (M€) [49]                                    | 709.6  |
|                                    | Reference power output (MW <sub>el,gross</sub> ) [49]                         | 580.2  |
| Calcium looping combustion process | Reference equipment capital cost (w/o CCU) (M€) [47]                          | 227.7  |
|                                    | Reference equipment capital cost (w/ CCU) (M€) [47]                           | 270.3  |
|                                    | Reference power output (MW <sub>el,gross</sub> ) [47]                         | 202.95 |
|                                    | Reference coal-fired power plant (-) [47]                                     | 0.67   |
| Scaling factor                     | Reactor volume (-) [48]                                                       | 0.67   |
|                                    | Heat input to the combustor (-) [48]                                          | 0.90   |
|                                    | Fraction of total system cost associated with heat transfer surfaces (-) [48] | 0.85   |
|                                    |                                                                               |        |
| Other economic parameters          | Variable cost as a fraction of total capital cost (%) [45,50]                 | 2.0    |
|                                    | Fixed cost as a fraction of total capital cost (%) [45,50]                    | 1.0    |
|                                    | Carbon tax (€/tCO <sub>2</sub> ) [45,50]                                      | 0.0    |
|                                    | Raw sorbent cost (€/t) [45,50]                                                | 6.0    |
|                                    | CO <sub>2</sub> transport and storage cost (€/tCO <sub>2</sub> ) [51]         | 7.0    |
|                                    | Coal price (€/t) [50,52]                                                      | 40.5   |
|                                    | Expected lifetime (years) [45,50]                                             | 25     |
|                                    | Project interest rate (%) [45,50]                                             | 8.78   |
|                                    | Capacity factor (%) [45,50]                                                   | 80     |

Finally, fixed and variable operating and maintenance costs are calculated as a fraction of total capital cost, while operating costs associated with fuel and sorbent consumption, and CO<sub>2</sub> storage, transport and emission are determined based on process simulation outputs using economic data from Table 2.

### 3.2 Techno-economic performance evaluation

The analysis of the thermodynamic performance of the CaLC process (Table 3) revealed that for the same heat input from coal combustion, its boiler thermal efficiency will be 2.0%<sub>HHV</sub> points higher than that of the conventional CFPP, implying a higher degree of fuel utilisation. The main reason behind such performance is a slightly higher degree of heat integration in the CaLC process that results from the heat carried by the sorbent purge stream being utilised for the fresh limestone make-up stream preheating. As a result, the heat losses are minimised, and thus the coal consumption is reduced. Furthermore, the heat from the clean gas leaving the reheater is utilised for feedwater heating, reducing the steam requirements in the feedwater heaters. No such integration is considered in the conventional CFPP to utilise the heat carried by the bottom ash stream and the waste heat from the flue gas.

**Table 3: Techno-economic performance indicators under initial design basis**

| Parameter                                                              | Conventional coal-fired power plant | Calcium looping combustion |          |
|------------------------------------------------------------------------|-------------------------------------|----------------------------|----------|
|                                                                        |                                     | Without CCU                | With CCU |
| Thermodynamic performance indicators                                   |                                     |                            |          |
| Heat input from coal combustion ( $\text{MW}_{\text{th}}$ )            | 1452.6                              | 1452.6                     | 1452.6   |
| Gross power output ( $\text{MW}_{\text{el}}$ )                         | 580.4                               | 583.8                      | 583.8    |
| Net power output ( $\text{MW}_{\text{el}}$ )                           | 552.7                               | 554.2                      | 507.1    |
| Net thermal efficiency ( $\%_{\text{HHV}}$ )                           | 38.0                                | 38.1                       | 34.9     |
| Boiler thermal efficiency ( $\%_{\text{HHV}}$ )                        | 85.2                                | 87.2                       | 87.2     |
| Specific coal consumption ( $\text{g/kW}_{\text{el}}\text{h}$ )        | 350.3                               | 349.3                      | 381.7    |
| Specific $\text{CO}_2$ emission ( $\text{g/kW}_{\text{el}}\text{h}$ )  | 796.8                               | 81.8                       | 89.4     |
| Specific $\text{NO}_x$ emissions ( $\text{g/kW}_{\text{el}}\text{h}$ ) | 0.6                                 | 0.4                        | 0.4      |
| Net efficiency penalty/gain ( $\%_{\text{HHV}}$ points)                | -                                   | -0.1                       | 3.1      |
| Economic performance indicators                                        |                                     |                            |          |
| Specific capital cost ( $\text{€/kW}_{\text{el,gross}}$ )              | 1222.6                              | 1547.5                     | 1837.1   |
| Levelised cost of electricity ( $\text{€/MW}_{\text{el}}\text{h}$ )    | 38.0                                | 47.0                       | 64.6     |
| $\text{CO}_2$ avoided cost ( $\text{€/tCO}_2$ )                        | -                                   | 12.5                       | 37.5     |

The CaLC process (without CCU) was found to be characterised by a net thermal efficiency of 38.1%<sub>HHV</sub> and low specific CO<sub>2</sub> emissions of 81.8 g/kW<sub>el</sub>h. This implies that the proposed process would yield a net efficiency gain of 0.1%<sub>HHV</sub> point compared to the conventional CFPP without CO<sub>2</sub> capture, and it will be characterised by approximately 90% lower specific CO<sub>2</sub> emissions. The thermodynamic performance of the CaLC with CCU, on the other hand, would be characterised by a net efficiency penalty of 3.1%<sub>HHV</sub> points and low specific CO<sub>2</sub> emissions of 89.4 g/kW<sub>el</sub>h. Furthermore, the proposed process would be characterised by comparable NO<sub>x</sub> emissions to that of the reference CFPP. Moreover, no SO<sub>x</sub> emissions are expected due to the high Ca:S ratios in the carbonator. Such performance is superior to the CaL retrofit scenario to the same CFPP, which was found to impose a net efficiency penalty of 5.8–7.9%<sub>HHV</sub> points depending on the design conditions and process configuration [16,53]. This analysis has shown that, considering both scenarios, the thermodynamic performance of the CaLC process compares favourably to:

- chemical looping combustion process that has been reported to have a net efficiency penalty below 4% points with CCU [32,54];
- combined calcium and chemical looping process for which the net efficiency penalty with CCU has been estimated to be 3.6–6.9% points [20,21];
- CaL with indirectly-heated calciner that has been reported to impose a net efficiency penalty of 3.6–6.3% points [20,21] (with CCU) and 1.5–3.5% points [26] (without CCU) in the retrofit or new-built scenario, respectively.

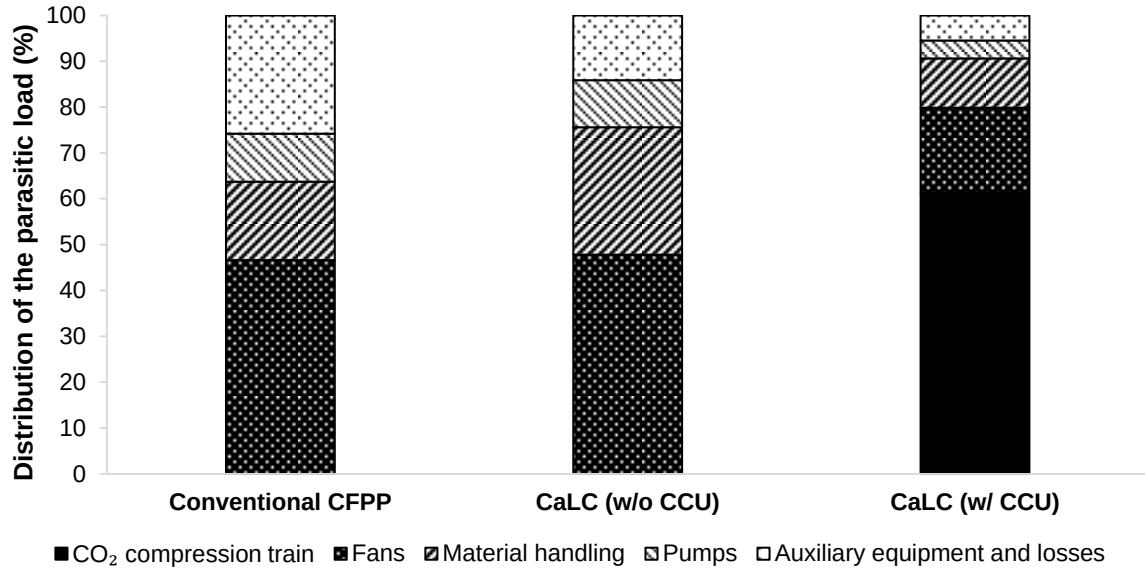


**Figure 2: Comparison of the parasitic load distribution**

Figure 2 reveals that similarly to the conventional CFPP, the main source of the parasitic load in the CaLC process (without CCU) are the fans (47.8%) increasing the pressure of the air and gas streams to overcome the pressure drop across the process equipment. On the contrary, the power requirement for the material handling equipment (27.8%) would be higher for the CaLC process, mainly due to the requirement to handle a larger amount of solids. Nevertheless, it is expected that the amount of solids to be handled would be one order of magnitude lower than in the dual-loop combined calcium and chemical looping process [20]. Importantly, the parasitic load associated with the auxiliary equipment and losses (14.1%) will be slightly smaller in the CaLC process due to the lack of a separate flue gas desulphurisation unit, which is mandatory for the conventional CFPP to meet the  $\text{SO}_x$  emission limits. Regardless of the flue gas desulphurisation taking place in the carbonator, the content of calcium sulphate in the purge stream is around 13%<sub>w</sub> that is comparable to values reported for CaL with direct oxy-combustion of coal in the calciner [55]. Finally, in the case of CaLC process with CCU, the power requirement corresponding to compression of  $\text{CO}_2$  prior to its transport and permanent storage

would account for more than 61% of the system's parasitic load. Therefore, further improvement in the thermodynamic performance of the CaLC process would mainly depend on reduction of the CCU power requirement, increasing the degree of heat integration, and utilising a power cycle with a higher thermal efficiency than that of the supercritical steam cycle, such as the supercritical CO<sub>2</sub> cycle [53].

Having proven the technical viability of the CaLC process, it is essential to benchmark its economic performance indicators with the figures for the conventional CFPP (Table 3). The specific capital cost of the CaLC process was estimated to be 1547.5 €/kW<sub>el,gross</sub> (without CCU) and 1837.1 €/kW<sub>el,gross</sub> (with CCU). These figures are close to the range estimated in other studies for the retrofit scenarios of both CaL (1250–1740 €/kW<sub>el,gross</sub>) [43,45,56] and CaL with indirectly-heated calciner (1791 €/kW<sub>el,gross</sub>) [25]. Yet, these specific capital costs are 26.6% and 50.3%, respectively, higher than the specific capital cost of the conventional CFPP without CO<sub>2</sub> capture (1222.6 €/kW<sub>el,gross</sub>) [49]. Moreover, the levelised costs of electricity associated with the CaLC process without and with CCU were estimated to be 47.0 €/MW<sub>el</sub>h and 64.6 €/MW<sub>el</sub>h, respectively, and the corresponding costs of CO<sub>2</sub> avoided were estimated as 12.5 €/tCO<sub>2</sub> and 37.5 €/tCO<sub>2</sub>, respectively. Therefore, the levelised cost of electricity is 23.6% and 70.0%, respectively, higher than that of the conventional CFPP. Yet, in both scenarios, the levelised cost of electricity and cost of CO<sub>2</sub> avoided are lower than that reported for the CaL retrofits (LCOE = 54.3–96 €/MW<sub>el</sub>h; AC = 28.9–58.3 €/tCO<sub>2</sub>) [45,48,57], chemical solvent scrubbing retrofits (LCOE = 65–75 €/MW<sub>el</sub>h; AC = 35–75 €/tCO<sub>2</sub>) [58–60], and chemical looping combustion (LCOE = 45–60 €/MW<sub>el</sub>h; AC = 16–55 €/tCO<sub>2</sub>) [32,61]. This implies that the CaLC process would become economically favoured over the conventional CFPP for a carbon tax higher than 12.5 €/tCO<sub>2</sub> in the case with CO<sub>2</sub> utilisation and 37.5 €/tCO<sub>2</sub> in

the case with permanent CO<sub>2</sub> storage, which is in the lower range of the predicted values for the carbon tax of 10–150 €/tCO<sub>2</sub> by 2050 [62,63].

### **3.3 Parametric study**

Performance of the CaLC process is directly dependent upon the specific coal consumption in the combustor, which, in turn, is directly dependent upon the amount of energy required to maintain its operating temperature. Under initial design conditions, the combustor operated at an equivalence ratio of 1.1 and operating temperature of 1000°C. Variation in the amount of excess air fed to the combustor (Figure 3) revealed that the techno-economic performance of CaLC improves with reduction in the equivalence ratio. This is a result of less heat required to preheat the combustion air to the combustor operating temperature. In addition, a lower flow rate of combustion air, and thus flue gas to be treated, allows more compact design of the CaLC process, reducing its capital cost. Importantly, increase of the equivalence ratio from 1.1 to 1.2 resulted in a net thermal efficiency reduction of 0.4% points and increase in the cost of CO<sub>2</sub> avoided of 2.7 €/tCO<sub>2</sub> (without CCU) and 3.7 €/tCO<sub>2</sub> (with CCU). Although a decrease in the CO emissions from 70.3 ppb<sub>v</sub> to 104.9 ppb<sub>v</sub> was observed, the former figure is still considerably lower than the allowable emission of around 8 ppm<sub>v</sub> [64].

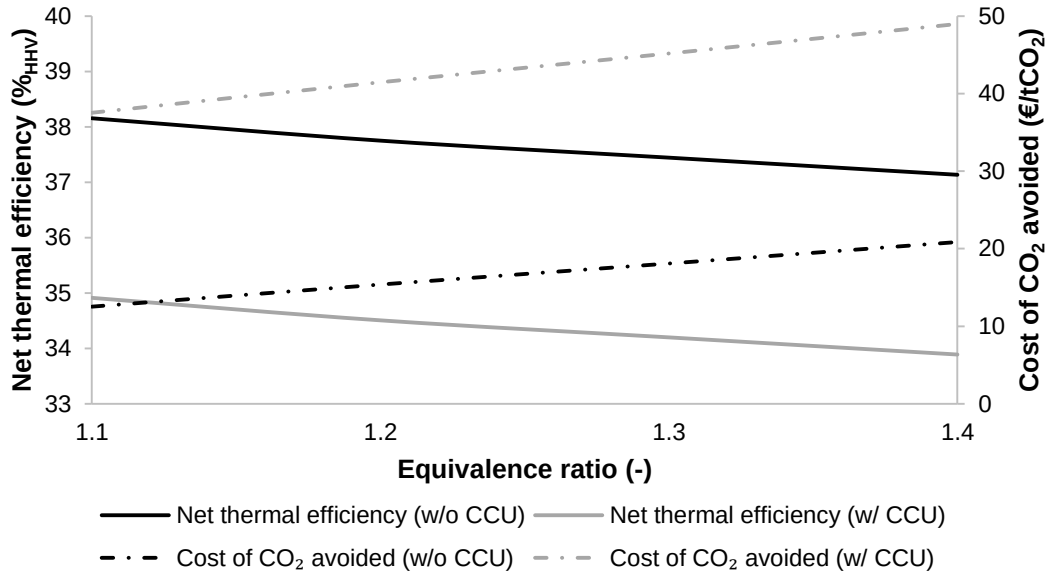
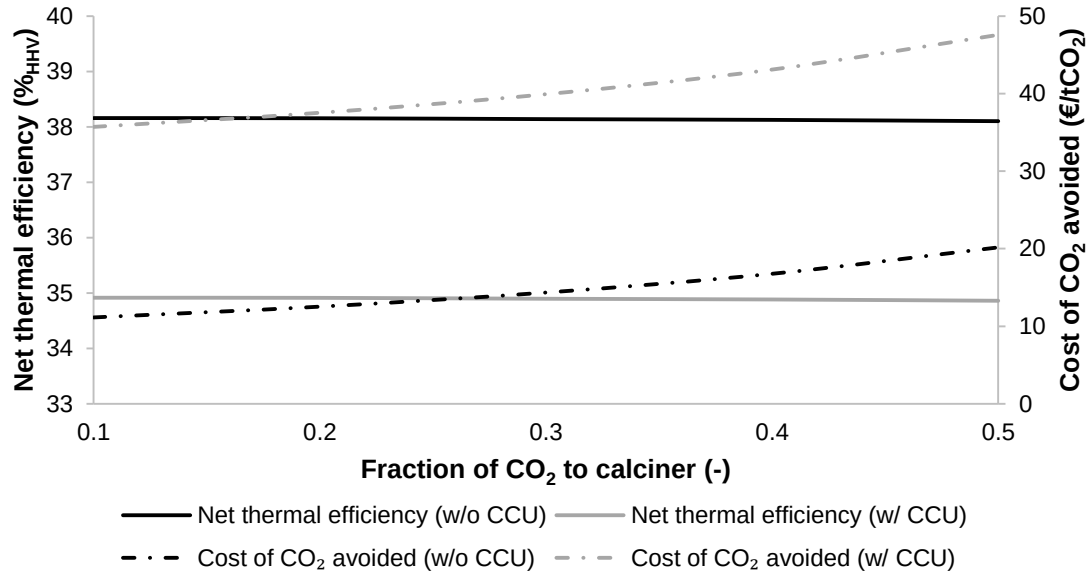
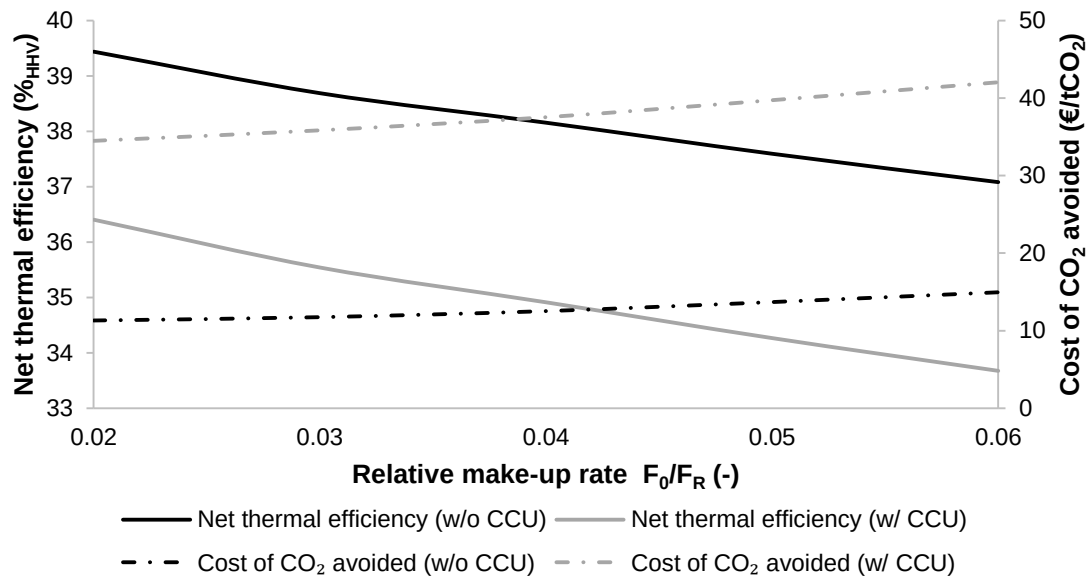


**Figure 3: Effect of the equivalence ratio on the process techno-economic performance**

The proposed CaLC process utilises part of the concentrated CO<sub>2</sub> stream as a fluidising medium to improve fluidisation and mixing of sorbent particles, and to avoid using steam that would affect the overall process efficiency. Under initial design conditions, 20% of the concentrated CO<sub>2</sub> was diverted to the calciner. Although variation of this parameter (Figure 4a) was found not to affect the net thermal efficiency of the CaLC process, its increase results in higher costs of CO<sub>2</sub> avoided. This can be primarily attributed to increased calciner capital cost with increasing volume flow rate of the fluidising medium. Therefore, on reduction of the fraction of concentrated CO<sub>2</sub> diverted to the calciner from 0.2 to 0.1, reductions in the cost of CO<sub>2</sub> avoided of 1.4 €/tCO<sub>2</sub> (without CCU) and 1.8 €/tCO<sub>2</sub> (with CCU) were observed.



a)



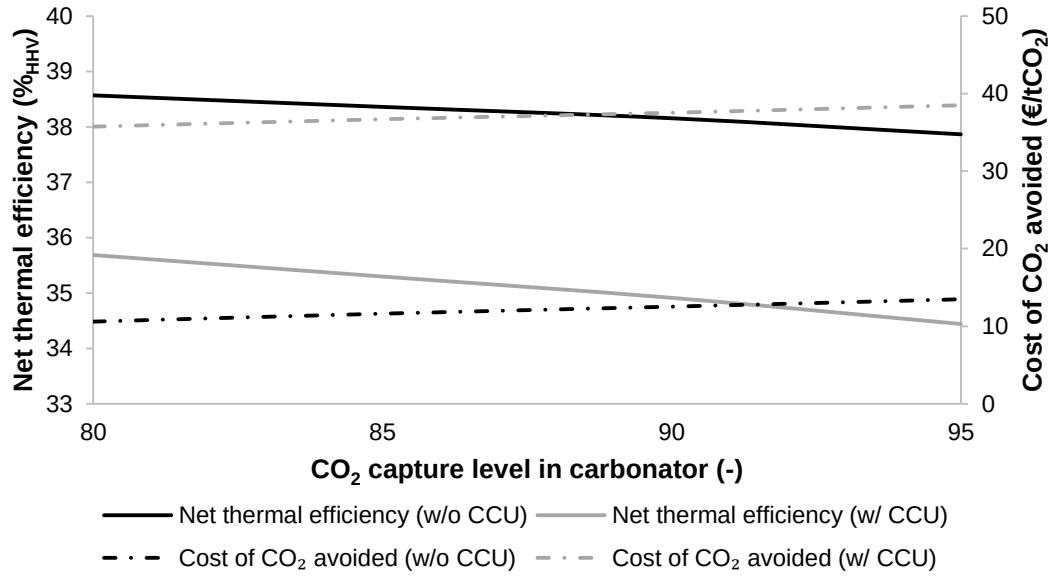
b)

**Figure 4: Effect of a) fraction of CO<sub>2</sub> to calciner and b) relative make-up rate on process techno-economic performance**

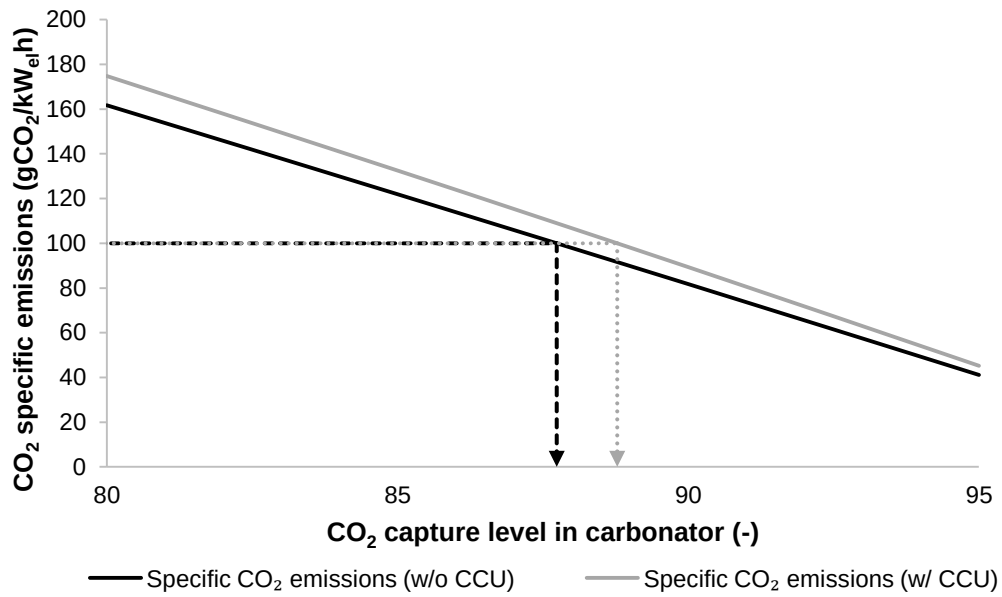
Importantly, the variation in the fresh limestone make-up rate, which is often represented by the ratio of fresh limestone make-up rate ( $F_0$ ) and sorbent looping rate ( $F_R$ ), was found to significantly affect the techno-economic performance of the CaLC process. Namely, the more fresh limestone that is fed to the system, the more energy is required for its preheating and calcination. As a result of higher sorbent

conversion in the carbonator, less solids are being circulated in the system, and thus less heat is available for recovery. This is reflected in a significant reduction in the net thermal efficiency of the CaLC process (Figure 4b). Furthermore, a corresponding increase in the cost of CO<sub>2</sub> avoided is observed, which results from a higher cost associated with more fresh limestone being utilised in, and the lower thermal efficiency of, the CaLC process. Nevertheless, on reduction of the relative make-up rate from the initial design value of 0.04 to 0.03, the net thermal efficiency increased by 0.5% points (without CCU) and 0.6% points (with CCU). Furthermore, the cost of CO<sub>2</sub> avoided was reduced by 0.8 €/tCO<sub>2</sub> (without CCU) and 1.7 €/tCO<sub>2</sub> (with CCU).

Finally, the evaluation of the process performance under initial design conditions indicated that the specific CO<sub>2</sub> emissions of the CaLC process were 81.8 g/kW<sub>el</sub>h (without CCU) and 89.4 g/kW<sub>el</sub>h (with CCU), implying that the carbonator can be operated with a CO<sub>2</sub> capture level below 90% while still meeting a specific CO<sub>2</sub> emission target of 100 g/kW<sub>el</sub>h. Reduction of the CO<sub>2</sub> capture level to 85% (Figure 5a) was found to increase the net thermal efficiency of the CaLC without and with CCU by 0.2% points and 0.4% points, respectively, as well as reduce the cost of CO<sub>2</sub> avoided by 0.9 €/tCO<sub>2</sub> and 0.8 €/tCO<sub>2</sub>, respectively. Yet, in this case the specific CO<sub>2</sub> emission was found to be 121.9 g/kW<sub>el</sub>h (without CCU) and 132.5 g/kW<sub>el</sub>h (with CCU), which is slightly above the targeted figure of 100 g/kW<sub>el</sub>h. Hence, the CO<sub>2</sub> capture level in the carbonator was optimised (Figure 5b), and was shown to be 87.7% (without CCU) and 88.7% (with CCU) for the CaLC process to meet the desired specific CO<sub>2</sub> emission target.



a)



b)

**Figure 5: Effect of CO<sub>2</sub> capture level in carbonator on process techno-economic performance**

### **3.4 Techno-economic performance evaluation under revised design basis**

The outcomes from the parametric study were used to revise the initial design basis of the CaLC process. To maximise the process performance, the relative fresh

sorbent make-up rate was reduced from 0.04 to 0.03, the fraction of the concentrated CO<sub>2</sub> diverted to the calciner was reduced from 0.2 to 0.1 and the CO<sub>2</sub> capture level in the carbonator was optimised to arrive at the desired specific CO<sub>2</sub> emissions of 100 g/kW<sub>el</sub>h.

**Table 4: Techno-economic performance indicators under revised design basis**

| Parameter                                                 | Conventional coal-fired power plant | Calcium looping combustion |          |
|-----------------------------------------------------------|-------------------------------------|----------------------------|----------|
|                                                           |                                     | Without CCU                | With CCU |
| Thermodynamic performance indicators                      |                                     |                            |          |
| Heat input from coal combustion (MW <sub>th</sub> )       | 1452.6                              | 1452.6                     | 1452.6   |
| Gross power output (MW <sub>el</sub> )                    | 580.4                               | 591.4                      | 591.4    |
| Net power output (MW <sub>el</sub> )                      | 552.7                               | 562.8                      | 517.8    |
| Net thermal efficiency (% <sub>HHV</sub> )                | 38.0                                | 38.7                       | 35.6     |
| Boiler thermal efficiency (% <sub>HHV</sub> )             | 85.2                                | 88.3                       | 88.3     |
| Specific coal consumption (g/kW <sub>el</sub> )           | 350.3                               | 344.0                      | 373.9    |
| Specific CO <sub>2</sub> emission (g/kW <sub>el</sub> )   | 796.8                               | 100.0                      | 100.0    |
| Specific NO <sub>x</sub> emissions (g/kW <sub>el</sub> h) | 0.6                                 | 0.4                        | 0.4      |
| Net efficiency penalty/gain (% <sub>HHV</sub> points)     | -                                   | -0.7                       | 2.4      |
| Economic performance indicators                           |                                     |                            |          |
| Specific capital cost (€/kW <sub>el,gross</sub> )         | 1222.6                              | 1485.9                     | 1773.0   |
| Levelised cost of electricity (€/MW <sub>el</sub> h)      | 38.0                                | 45.0                       | 61.7     |
| CO <sub>2</sub> avoided cost (€/tCO <sub>2</sub> )        | -                                   | 10.0                       | 33.9     |

As a result (Table 4), the net thermal efficiency of the CaLC process (without CCU) increased from 38.1%<sub>HHV</sub> to 38.7%<sub>HHV</sub>, resulting in 0.7%<sub>HHV</sub> point net efficiency gain compared to the conventional CFPP, while the specific CO<sub>2</sub> emission target of 100 g/kW<sub>el</sub>h is met. Such performance is superior to the CFPP based on chemical looping combustion, which was reported to yield no net efficiency penalty [54]. In addition, the amount of solids to be handled in the CaLC process would be, on average, 2.4 times lower compared to the system based on chemical looping combustion, implying lower power requirements for solid handling. This is because the average mass flow rate ratios of solids circulated between the reactors and the steam entering the HP turbine in the CaLC process proposed in this study and the

CFPP based on chemical looping combustion analysed by Spinelli et al. [54] are 3.7 and 8.7, respectively<sup>†</sup>. Furthermore, the CaLC process with CCU would impose a small net efficiency penalty of 2.4% points, which is similar to the net efficiency penalty reported for the CaL with an indirectly-heated calciner in the retrofit scenario without CCU [26]. It needs to be stressed that the net thermal efficiency of the CaLC process is comparable to the conventional CFPP, superior to the CFPP based on chemical looping combustion, and is considerably higher than in the CO<sub>2</sub> retrofit scenarios to the CFPPs reported in the literature.

The capital cost of the CaLC process under the revised design basis was reduced to 1485.9 €/kW<sub>el,gross</sub> (without CCU) and 1773.0 €/kW<sub>el,gross</sub> (with CCU), which are within the range of values reported in other studies [25,43,45,56]. Importantly, the levelised cost of electricity for the CaLC process without CCU is only 7.0 €/MW<sub>el</sub>h higher than that of the conventional CFPP and the cost of CO<sub>2</sub> avoided is 10.0 €/tCO<sub>2</sub>. For the CaLC process with CCU, the cost of CO<sub>2</sub> avoided was estimated to be 33.9 €/tCO<sub>2</sub>. As the carbon tax has recently fluctuated between 4.3 and 8.8 €/tCO<sub>2</sub> [65], and it is predicted to reach 10–150 €/tCO<sub>2</sub> by 2050 [62,63], it is expected that the CaLC process (both without and with CCU) would become more economically favoured than the conventional CFPP.

Considering the fact that the CaLC process is based on the state-of-the-art CaL (TRL=6) and the CaL with indirectly heated calciner (TRL=3) processes, for which the technology readiness level is higher than that of chemical looping combustion (TRL=2) [66], it can be seen as a high-efficiency low-emission power generation

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<sup>†</sup> Estimation of the average mass flow rate ratios of solids circulated between the reactors and the steam entering the HP turbine is available in Supporting Information.

technology that could become commercially available earlier than the CFPPs based on chemical looping combustion. Further improvement in the techno-economic performance of the CaLC process can be achieved mainly via increasing the degree of heat integration, and utilising a power cycle of higher thermal efficiency than that of the supercritical steam cycle, such as the supercritical CO<sub>2</sub> cycle [53], the specific capital cost of which has been estimated to be up to 27% lower than that for the supercritical steam cycle operating in the same envelope [67].

## 4 CONCLUSIONS

This study intended to establish a new class of high-temperature solid looping combustion technologies for high-efficiency low-emission power generation by proposing a novel calcium looping combustion (CaLC) process, the techno-economic performance of which compares favourably with conventional CFPPs without CO<sub>2</sub> capture. The techno-economic performance of the CaLC process with the concentrated CO<sub>2</sub> stream being sent either for utilisation (without CCU) or permanent storage (with CCU) was benchmarked against the conventional 580 MW<sub>el</sub> CFPP. The evaluation under the initial design basis has indicated that the CaLC process has a higher boiler thermal efficiency (87.2%<sub>HHV</sub>) than the conventional power boiler (85.2%<sub>HHV</sub>). Such performance results from a slightly higher degree of heat integration in the CaLC process as the heat carried by the sorbent purge stream is utilised for the fresh limestone make-up stream preheating. The net thermal efficiency of the proposed system without CCU was 0.1%<sub>HHV</sub> points higher, while with CCU was 3.1% lower, than that of the conventional CFPP. The associated costs of CO<sub>2</sub> avoided were 12.5 €/tCO<sub>2</sub> and 37.5 €/tCO<sub>2</sub>, respectively. Having revised the design basis using the findings from the parametric study, the net thermal efficiency of the CaLC without CCU was 0.7%<sub>HHV</sub> points higher, while with CCU was 2.4%<sub>HHV</sub>

points lower than that of the conventional CFPP. Therefore, the thermodynamic performance of the CaLC process would be comparable to that of the conventional CFPP, while its emissions would meet the specific CO<sub>2</sub> emission target of 100 g/kW<sub>el</sub>h. Importantly, the cost of CO<sub>2</sub> avoided for the CaLC process under the revised design basis was estimated to be 10.0 €/tCO<sub>2</sub> (without CCU) and 33.9 €/tCO<sub>2</sub> (with CCU). With the carbon tax currently varying between 4.3 €/tCO<sub>2</sub> and 8.8 €/tCO<sub>2</sub>, and being expected to rise to 10–150 €/tCO<sub>2</sub>, it is expected that the CaLC process could become more economically favoured than the conventional CFPP in the short- to mid-term.

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## NOMENCLATURE

|                  |                                                                                                              |                                      |
|------------------|--------------------------------------------------------------------------------------------------------------|--------------------------------------|
| $AC$             | Cost of CO <sub>2</sub> avoided                                                                              | €/tCO <sub>2</sub>                   |
| $C$              | Capital cost                                                                                                 | €/kW <sub>el</sub>                   |
| $C_0$            | Reference capital cost of                                                                                    | €/kW <sub>el</sub>                   |
| $CF$             | Capacity factor                                                                                              | -                                    |
| $e_{CO_2}$       | Specific CO <sub>2</sub> emission                                                                            | gCO <sub>2</sub> /kW <sub>el</sub> h |
| $EP$             | Net efficiency penalty                                                                                       | % <sub>HHV</sub> points              |
| $F_0$            | Fresh limestone make-up rate                                                                                 | kmol/s                               |
| $FCF$            | Fixed charge factor                                                                                          | -                                    |
| $FOM$            | Fixed operating and maintenance cost                                                                         | €                                    |
| $F_R$            | Sorbent looping rate                                                                                         | kmol/s                               |
| $LCOE$           | Levelised cost of electricity                                                                                | €/MW <sub>el</sub> h                 |
| $\dot{m}_{CO_2}$ | Rate of CO <sub>2</sub> emission                                                                             | kg/s                                 |
| $SCF$            | Specific fuel cost                                                                                           | €/MW <sub>ch</sub>                   |
| $SF, Q$          | Scaling factor for reactor heat input                                                                        | -                                    |
| $SF, V$          | Scaling factor for reactor volume                                                                            | -                                    |
| $TCR$            | Total capital requirement                                                                                    | €                                    |
| $\dot{Q}_0$      | Reference heat input                                                                                         | MW <sub>th</sub>                     |
| $\dot{Q}_{comb}$ | Heat input to the combustor                                                                                  | MW <sub>th</sub>                     |
| $\dot{Q}_{fuel}$ | Chemical energy input from fuel combustion                                                                   | MW <sub>ch</sub>                     |
| $\dot{Q}_{sc}$   | Heat transferred to the steam cycle                                                                          | MW <sub>th</sub>                     |
| $V$              | Volume of reactors                                                                                           | m <sup>3</sup>                       |
| $V_0$            | Reference volume of reactor                                                                                  | m <sup>3</sup>                       |
| $VOM$            | Variable operating and maintenance cost                                                                      | €/MW <sub>el</sub> h                 |
| $\dot{W}_{net}$  | Net power output of the integrated system                                                                    | MW <sub>el</sub>                     |
| $\alpha$         | Fraction of the total cost of a circulating fluidised bed reactor associated with the heat transfer surfaces | -                                    |
| $\eta_{th}$      | Net thermal efficiency                                                                                       | -                                    |
| $\eta_b$         | Boiler thermal efficiency                                                                                    | -                                    |

## ABBREVIATIONS

|      |                                  |
|------|----------------------------------|
| CaL  | Calcium looping                  |
| CaLC | Calcium looping combustion       |
| CCS  | Carbon capture and storage       |
| CCU  | CO <sub>2</sub> compression unit |
| CFPP | Coal-fired power plant           |
| HP   | High-pressure                    |
| IP   | Intermediate-pressure            |
| LP   | Low-pressure                     |



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**Hanak, Dawid P.; Manovic, Vasilije**

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