Calcium looping with supercritical CO$_2$ cycle for
decarbonisation of coal-fired power plant

Dawid P. Hanak*, Vasilije Manovic

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Combustion and CCS Centre, Cranfield University,
Bedford, Bedfordshire, MK43 0AL, UK

Corresponding author*: Dawid Hanak
d.p.hanak@cranfield.ac.uk
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Abstract

State-of-the-art integration scenarios of calcium looping (CaL), which is an emerging CO\textsubscript{2} capture technology, assume that excess heat is used to raise steam for the steam cycle and result in a net efficiency penalty of 6.0–8.0\% points. In this study, a concept using the supercritical CO\textsubscript{2} cycle (s-CO\textsubscript{2}) instead of the conventional steam cycle is proposed. Retrofit of CaL with recompression s-CO\textsubscript{2} cycle to the 580 MW\textsubscript{el} coal-fired power plant was found to result in a net efficiency penalty of 6.9\%\textsubscript{HHV} points. This is 1\%\textsubscript{HHV} point lower than that for the same system linked with the steam cycle having the same turbine inlet conditions (593.3°C/242.3 bar). A further reduction of the net efficiency penalty to 5.8\%\textsubscript{HHV} points was achieved through considering a pump instead of a first CO\textsubscript{2} compression stage and increasing the turbine inlet temperature to 620°C and pressure to 300 bar. As the s-CO\textsubscript{2} cycle’s specific capital cost is up to 27\% lower than that of the equivalent steam cycle, CaL with s-CO\textsubscript{2} cycle is a viable option for the coal-fired power plant decarbonisation. Moreover, it can be expected that this cycle can be successfully implemented in other high-temperature looping cycles, such as chemical looping combustion.

Key Words: Calcium looping, carbon capture, coal-fired power plant, supercritical CO\textsubscript{2} cycle, recompression Brayton cycle, efficiency penalty reduction
1 INTRODUCTION

The power generation sector is one of the key sectors that need to be decarbonised by 2050 to meet the greenhouse gas emission reduction target and to mitigate drastic climate change [1–4]. It has been predicted that its near-complete decarbonisation would account for approximately 28% of the total CO₂ emission reduction in 2025 [5]. Regardless of that, coal-fired power plants are currently responsible for more than 70% of the total CO₂ emissions from the power sector and their share in the energy portfolio is predicted to decrease only by around 6%, for example, from around 40% [5,6] in 2013 to 34% in 2040 [6]. On the other hand, a 5% increase in the renewable energy sources share is predicted within this time frame [6]. Yet, large-scale deployment of renewables requires more backup capacity to make up for their intermittency, introducing additional costs. As a result, coal-based power generation remains more attractive for investors and is expected to remain the key power source in the future energy portfolio [7]. Therefore, carbon capture and storage (CCS) is expected to act, at least, as a bridge to the clean power generation future [8].

Regardless of the recent progress in oxy-combustion and chemical solvent scrubbing post-combustion CO₂ capture technologies [9,10], the efficiency penalty associated with integration of these systems to coal-fired power plants is predicted to be 7.0–12.5% points [11–16], which would increase the cost of electricity by at least 60% [17–20]. Therefore, emerging technologies, which offer more competitive performance of the retrofitted system, are being developed [21]. Calcium looping (CaL), which is based on the reversible carbonation reaction of lime, has been proposed as a viable and efficient process for decarbonisation of coal-fired power plants [22]. A recent review [23] revealed that the efficiency penalty associated with CaL is projected to be within 6.0–8.0% points, depending on the level of heat integration. The main reason
behind the superior performance of CaL compared to the more mature CO₂ capture technologies, is its high-temperature operation and high-grade heat availability [22,24,25].

The state-of-the-art CaL integration scenarios assume that high-grade heat is recovered to generate steam at a high pressure, which is then used to generate power in the conventional steam cycle based on the Rankine cycle. Yet, it is worth highlighting that CaL operates within the temperature range of 600–950°C [23], which is similar to the operating temperatures of the natural gas turbine bottoming cycle (600°C) [26], high-temperature gas-cooled nuclear reactors (850–1000°C) [27], sodium-cooled fast nuclear reactors (500–550°C) [28,29], helium-cooled fusion nuclear reactors (500°C) [30] and concentrating solar power plants (800–900°C) [31,32] – technologies for which alternative thermodynamic cycles, such as the Brayton cycle using He or supercritical CO₂ (s-CO₂), have been investigated.

Compared to the conventional supercritical steam cycle based on the Rankine cycle, the He and s-CO₂ cycles can achieve higher cycle efficiencies that can be as high as 50-60%, depending on the turbine inlet conditions and the cycle architecture [31–34]. Dostal et al. [34] have shown that the s-CO₂ cycle outperforms the equivalent He cycle at the same turbine inlet temperature. This is mostly because of near-critical-point operation of the CO₂ compressors resulting in the compression power requirement being only around 30% of the turbine power output. Also, due to the high density and low heat capacity of supercritical CO₂, the size of the turbomachinery is considerably smaller than that for the steam cycle and the He cycle. In addition, the layout of the system based on the Brayton cycle is simpler than that for the steam cycle [34–37].

All of these features of the s-CO₂ cycle are reflected in the specific capital cost ($/MWₑₑ), which for this cycle has been estimated to be up to 27% lower than for the
superheated steam cycle and 11% lower than the equivalent He cycle [34], making the s-CO$_2$ cycle a technically and economically viable option for future power generation systems. This is also reflected in a wide spectrum of investigated applications of the s-CO$_2$ cycle that vary from low-grade waste heat recovery systems [38], fuel cell hybrid systems [39] and liquefied natural gas power and refrigeration systems [40,41], through to solar [31,32,42], nuclear [27–30,34,38,43] and even coal-fired power plants [44]. The aforementioned qualities make the s-CO$_2$ cycle worth considering as an alternative to the conventional Rankine steam cycle [35].

Reduction of the efficiency and economic penalties associated with CO$_2$ capture is required for the industrial deployment of these technologies. CaL, as an emerging CO$_2$ capture technology, has already been proven as a viable alternative to mature technologies for decarbonisation of coal-fired power plants. Yet, the state-of-the-art integration scenarios suggest that the excess high-grade heat from the CaL plant can be utilised in the primary or the secondary steam cycle for power generation [23]. Due to higher cycle efficiency and lower specific capital cost associated with the s-CO$_2$ cycle compared to the steam cycle, the concept of the CaL plant with s-CO$_2$ cycle for decarbonisation of a coal-fired power plant is proposed in this study. For the purpose of this study, the s-CO$_2$ process model was developed and then adapted to the existing CaL process model [45]. The performance of the proposed retrofit scenario is first compared with the CaL plant with the supercritical reheated steam cycle reported by Hanak et al. [45]. Then, the impact of variations of the s-CO$_2$ hardware and the turbine inlet conditions on the net thermal efficiency of the retrofitted system are investigated. Finally, the optimum design is proposed based on sensitivity studies and further improvements in the retrofitted system performance are identified.
2 PROCESS MODEL DEVELOPMENT

2.1 Process description

This study analyses a scenario in which the CaL plant, which comprises two interconnected fluidised bed reactors operating at atmospheric pressure, is retrofitted to a 580 MW \textsubscript{el} coal-fired power plant to achieve a total CO\textsubscript{2} capture level of 90%. In this system, CO\textsubscript{2} from the flue gas is removed in the carbonator, which operates at 650°C, through the exothermic reaction with CaO. The saturated sorbent (CaCO\textsubscript{3}) is then regenerated at a high temperature (900°C) in the calciner, which is ensured through oxy-combustion of coal. The high-temperature operation of the CaL plant allows for high-grade heat recovery from the carbonator and the process streams. The high-grade heat is utilised to generate an additional amount of power in the s-CO\textsubscript{2} cycle, which is evaluated in this study as an alternative to the supercritical steam cycle considered thus far in the literature [22–24,46–53].

![Figure 1: Scheme of the recompression supercritical CO\textsubscript{2} (s-CO\textsubscript{2}) cycle](image)

The s-CO\textsubscript{2} cycle, which was introduced by Feher [36], is a closed-loop Brayton cycle that is highly recuperative and operates entirely above CO\textsubscript{2} critical pressure [28], [36,38]. The heat capacity of supercritical CO\textsubscript{2} varies substantially with pressure [29],
and thus the simple s-CO$_2$ cycle suffers from large irreversibility in the recuperator caused by the heat capacity imbalance between its hot and cold sides [35]. This causes a pinch point problem in the recuperator [31,35] that is reflected in the minimum temperature approach occurring inside the heat exchanger [34]. In the recompression s-CO$_2$ cycle (Figure 1), which is considered in this study, this issue is resolved through recompressing part of the CO$_2$ stream without heat rejection to compensate the heat capacity difference in a low-temperature recuperator (LTR) [29]. As a result of increased LTR effectiveness, the hot stream enters a high-temperature recuperator (HTR) at a higher temperature. This, in turn, increases the temperature of the hot stream leaving the HTR and thus the average temperature of heat addition, leading to higher cycle efficiency [28].

### 2.2 Supercritical coal-fired power plant

A process model of a 580 MW$_{el}$ supercritical coal-fired power plant, which is considered as a benchmark in this study, was developed in Aspen Plus and described in detail by Hanak et al. [25,54]. The prediction of this model was found to be in close agreement with data provided in the revised NETL report [55], confirming the efficacy of the modelling approach and high fidelity of the model.

The coal-fired power plant model comprises three sub-models: supercritical boiler and flue gas treatment train (NO$_x$, SO$_x$ and fly ash) models, both modelled using Peng–Robinson–Boston–Mathias (PR–BM) equation of state, as well as a steam cycle model that has been thermodynamically described with steam tables (STEAMNBS). Coal combustion is modelled using solid modelling features recommended by AspenTech [56]. The boiler heat exchange sections, which include the primary, secondary, and reheat superheaters, as well as the economiser, are modelled using $HEATX$ block with pressure drops and temperature levels set based on the revised NETL report [55].
Steam turbines are modelled as individual turbine sections using COMPR block, hence the high-, intermediate- and low-pressure cylinders are further divided into the required steam turbine sections. The isentropic efficiency was estimated to match the desired discharge temperature of each section, which was provided in the revised NETL report [55]. The condenser is modelled as HEATX block and is assumed to operate at a fixed pressure of 0.069 bar, which corresponds to a condensation temperature of 38.7°C. The performance of the modelled coal-fired power plant is characterised by gross power output of 580.4 MW_{el}, net thermal efficiency of 38.5%_{HHV} and other key performance parameters estimated by Hanak et al. [25] and presented in Table 1.

Table 1: Reference coal-fired power plant and CaL retrofit (supercritical steam cycle) operating specification

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Reference coal-fired power plant [25]</th>
<th>CaL retrofit (supercritical steam cycle) [45]</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>System performance indicators</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gross power output (MW_{el})</td>
<td>580.4</td>
<td>1023.3</td>
</tr>
<tr>
<td>Net power output (MW_{el})</td>
<td>552.7</td>
<td>799.9</td>
</tr>
<tr>
<td>Net thermal efficiency (%_{HHV})</td>
<td>38.5</td>
<td>30.6</td>
</tr>
<tr>
<td>Specific coal consumption (g/kW_{el}h)</td>
<td>350.3</td>
<td>438.2</td>
</tr>
<tr>
<td><strong>Calcium looping specification</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Limestone make-up rate (F_{0}/F_{R}) (-)</td>
<td>-</td>
<td>0.04</td>
</tr>
<tr>
<td>Carbonator temperature (°C)</td>
<td>-</td>
<td>650</td>
</tr>
<tr>
<td>Calciner temperature (°C)</td>
<td>-</td>
<td>900</td>
</tr>
<tr>
<td>Oxygen content in calciner fluidising gas (%_{vol,wet})</td>
<td>-</td>
<td>40</td>
</tr>
<tr>
<td>Excess oxygen (%_{vol,dry})</td>
<td>-</td>
<td>2.5</td>
</tr>
<tr>
<td><strong>Primary and secondary steam cycle specification</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Live steam pressure (bar)</td>
<td>242.3</td>
<td></td>
</tr>
<tr>
<td>Reheated steam pressure (bar)</td>
<td>45.2</td>
<td></td>
</tr>
<tr>
<td>Intermediate-/low-pressure crossover pressure (bar)</td>
<td>9.3</td>
<td></td>
</tr>
<tr>
<td>Condenser pressure (bar)</td>
<td>0.069</td>
<td></td>
</tr>
<tr>
<td>Live and reheated steam temperature (°C)</td>
<td>593.3</td>
<td></td>
</tr>
</tbody>
</table>
2.3 Calcium looping plant with supercritical steam cycle

The process model of the CaL plant, which is modelled using SOLIDS property method, with a supercritical steam cycle was developed in Aspen Plus and described in detail in Hanak et al. [45]. The prediction of the CaL model showed good agreement with data available in the literature [57–60].

In the CaL model, the carbonation process is modelled using the \textit{RSTOIC} block in conjunction with the maximum average sorbent conversion model derived by Rodríguez et al. [61] and presented in Eq. (1). The maximum average conversion in this model is a function of the carbonation ($f_{\text{carb}}$) and calcination extent ($f_{\text{calc}}$), sorbent characteristics ($a_1, a_2, f_1, f_2, b$), which were estimated based on experimental data from the 1.7 MW\textsubscript{th} INCAR-CSIC pilot plant [57], fresh limestone make-up ($F_0$) and solids looping rate ($F_R$). The carbonation conversion and the CO\textsubscript{2} capture level, which are used to determine the solids looping rate, are estimated using Eq. (2) and Eq. (3), respectively.

\[
x_{\text{ave}} = \left( F_0 + F_R \frac{F_0(1-f_{\text{calc}})}{F_0 + F_R f_{\text{calc}}} \right) f_{\text{calc}} \left[ \frac{a_1 f_1^2}{F_0 + F_R f_{\text{carb}} f_{\text{calc}} (1-f_1)} + \frac{a_2 f_2^2}{F_0 + F_R f_{\text{carb}} f_{\text{calc}} (1-f_2)} + \frac{b}{F_0} \right] \tag{1}
\]

\[
\Delta X_{\text{carb}} = f_{\text{calc}} f_{\text{carb}} x_{\text{ave}} \tag{2}
\]

\[
E_{\text{CO}_2} = \frac{F_R}{F_{\text{CO}_2}} \Delta X_{\text{carb}} \tag{3}
\]

The calcination process, which aims at regeneration of the saturated sorbent, is modelled using \textit{RGibbs} block that determines the chemical and phase equilibrium through the Gibbs' free energy minimisation at a given temperature. The equilibrium limits of the carbonation and calcination processes are accounted for using the correlation derived by Baker [62] to estimate the equilibrium partial pressure of CO\textsubscript{2} in the gas streams leaving the reactors. Finally, the CaL plant model comprises the auxiliary units, such as conventional air separation unit (ASU) and CO\textsubscript{2} compression
unit (CCU), for which a detailed modelling approach is widely described in the literature [45,54,63–66].

In the retrofitted system operating with the characteristics presented in Table 1, which were identified in Hanak et al. [45], the high-grade heat from the CaL plant was utilised to generate high-pressure steam, which has the same parameters as the supercritical boiler (593.3°C/593.3°C/242.3 bar), for the secondary steam cycle of similar operating conditions to the primary steam cycle. Both the heat recovery steam generator (HRSG) and the secondary steam cycle were modelled using the same approach as adapted for the coal-fired power plant model. The analysis performed by Hanak et al. [45] revealed that retrofit of the CaL plant with supercritical steam cycle to the reference coal-fired power plant increased the net power output of the retrofitted system by 44.7% and resulted in 7.9% HHV points drop in its net efficiency (Table 1). The performance of the CaL plant with the supercritical steam cycle will be used to assess if application of the s-CO₂ cycle to recover the high-grade heat from the CaL plant will bring performance benefits.

2.4 Supercritical CO₂ cycle

2.4.1 Model description and verification

A process model of the recompression s-CO₂ cycle has been developed in Aspen Plus based on data from Moisseytsev and Sienicki [29], who analysed the integration of the s-CO₂ cycle with the reference 96 MWₑₑ sodium-cooled fast reactor. One of the main technical challenges of the s-CO₂ cycle is estimation accuracy of the turbomachinery efficiencies [44], as these units are still under development and only a few have been tested at bench- and pilot-scale [42,67]. In the open literature, the performance of the CO₂ turbine is usually represented using its isentropic efficiency, the typical value of
which is between 87.0% and 93.4% [28,31,34,43,68,69]. Similarly, the performance of the main and the recompression compressors is represented by isentropic efficiency of 85.0–95.5% [28,31,34,43,44].

**Table 2: Reference recompression supercritical CO\(_2\) cycle operating parameters**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Main compressor discharge pressure (bar)</td>
<td>200.0</td>
</tr>
<tr>
<td>Recompression compressor discharge pressure (bar)</td>
<td>199.6</td>
</tr>
<tr>
<td>Turbine discharge pressure (bar)</td>
<td>77.3</td>
</tr>
<tr>
<td>Cycle lowest pressure (bar)</td>
<td>74.0</td>
</tr>
<tr>
<td>Cooler outlet temperature (°C)</td>
<td>31.3</td>
</tr>
<tr>
<td>Fraction of CO(_2) to recompression (%)</td>
<td>29.0</td>
</tr>
<tr>
<td>Turbine isentropic efficiency (%)</td>
<td>93.0</td>
</tr>
<tr>
<td>Compressor isentropic efficiency (%)</td>
<td>90.0</td>
</tr>
<tr>
<td>Mechanical efficiency of the rotational machinery (%)</td>
<td>99.0</td>
</tr>
<tr>
<td>Generator efficiency (%)</td>
<td>98.5</td>
</tr>
</tbody>
</table>

**Table 3: Benchmark of the supercritical recompression CO\(_2\) cycle stream data with Moisseytsev and Sienicki [29]**

<table>
<thead>
<tr>
<th>Stream</th>
<th>Temperature (°C)</th>
<th>Pressure (bar)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Literature</td>
<td>Model</td>
</tr>
<tr>
<td>1</td>
<td>31.3</td>
<td>31.3</td>
</tr>
<tr>
<td>2</td>
<td>84.4</td>
<td>82.8</td>
</tr>
<tr>
<td>3</td>
<td>171.8</td>
<td>171.8</td>
</tr>
<tr>
<td>4</td>
<td>323.3</td>
<td>323.3</td>
</tr>
<tr>
<td>5</td>
<td>471.8</td>
<td>471.5</td>
</tr>
<tr>
<td>6</td>
<td>362.3</td>
<td>361.8</td>
</tr>
<tr>
<td>7</td>
<td>190.7</td>
<td>189.0</td>
</tr>
<tr>
<td>8</td>
<td>90.2</td>
<td>87.7</td>
</tr>
</tbody>
</table>

Using the parameters presented in Table 2, which were taken from Moisseytsev and Sienicki [29] and Le Moullec [44], the Lee-Kesler-Plocker equation of state [44], the HEATX blocks to represent performance of the LTR, HTR and cooler, and the COMPR blocks to represent performance of the CO\(_2\) turbines and compressors, the net efficiency of the cycle presented in Figure 1 is estimated to be 39.6%, which is 0.8% points lower than that of the reference study [29]. The main reason for such deviation
is the fact that the isentropic efficiencies for the \( \text{CO}_2 \) turbine and compressors were not provided in the reference study [29] and thus were taken from Le Moullec [44], who used the same reference system in his study. This is reflected in small deviations in prediction of the temperatures for the steam leaving the main compressor and the \( \text{CO}_2 \) turbine (Table 3). Nevertheless, the model prediction was found to be in good agreement with the literature data as the highest relative error was 2.8%.

### 2.4.2 Adaptation of supercritical \( \text{CO}_2 \) cycle to calcium looping

A large fraction of the high-grade heat needs to be recovered from the flue gas and pure \( \text{CO}_2 \) streams leaving the CaL plant. The recompression s-CO\(_2\) cycle shown in Figure 1 cannot, therefore, be directly integrated with CaL because the sensible heat of the process streams below 500°C would not be utilised in the system, leading to high energy losses. The same issue was encountered by Le Moullec [44], who analysed the implementation of the recompression s-CO\(_2\) cycle in place of the steam cycle in the coal-fired power plant with an amine scrubbing \( \text{CO}_2 \) capture plant. This study showed that the low-grade heat can be recovered from the process streams by extracting part of the \( \text{CO}_2 \) stream leaving the main \( \text{CO}_2 \) compressor at low temperature and returning it after the HTR. The same approach is applied in this study (Figure 2) and the low-grade heat from the clean flue gas stream is recovered by cooling it to 60.0°C using around 15–17% of the \( \text{CO}_2 \) flow leaving the main compressor at 44.4°C. Moreover, heat carried with the \( \text{CO}_2 \) stream at 900°C is first used to superheat the \( \text{CO}_2 \) stream to the desired high-pressure turbine inlet temperature, and then to preheat the flue gas and the \( \text{O}_2 \) streams entering the CaL plant.

To maximise the CaL excess heat utilisation and the efficiency of the recompression s-CO\(_2\) cycle, a single reheat and a single intercooling stage are considered in the base configuration. In this configuration, the high-pressure and the intermediate-pressure
turbine inlet temperature is 593.3°C. In addition, the high-pressure turbine inlet pressure is set as 242.3 bar and both turbine stages operate with constant pressure ratio. Such assumptions for the base configuration allow its direct comparison with the reference configuration, in which the excess heat from CaL is utilised in the supercritical reheated steam cycle. Design of the heat exchanger network is based on the assumption that the minimum temperature approach of the heat exchangers is 10°C [70], with exception of recuperators and superheaters/reheaters for which the minimum temperature approach of 5°C [31], [32] and 25°C [70], respectively, is used. Finally, the reported values for the mechanical efficiency of the turbomachinery and the electric efficiency of the generator vary between 99.6–100.0% [28,70] and 97.5%–98.5% [29,55,70], respectively. In this study the mechanical efficiencies of 99.8% and 99.6% are selected for CO₂ turbines and compressors, respectively, while the electric efficiency of 98.0% is chosen for the generator. The key design parameters for the recompression s-CO₂ cycle adapted to the CaL plant are summarised in Table 4.

**Table 4: Design basis for the recompression supercritical CO₂ cycle**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Isentropic efficiency of CO₂ turbines (%)</td>
<td>93.0</td>
</tr>
<tr>
<td>Isentropic efficiency of main compressors (%)</td>
<td>90.0</td>
</tr>
<tr>
<td>Isentropic efficiency of recompression compressor (%)</td>
<td>90.0</td>
</tr>
<tr>
<td>Mechanical efficiency of CO₂ turbines (%)</td>
<td>99.8</td>
</tr>
<tr>
<td>Mechanical efficiency of compressors (%)</td>
<td>99.6</td>
</tr>
<tr>
<td>Electric generator efficiency (%)</td>
<td>98.0</td>
</tr>
<tr>
<td>Reheater and superheater minimum temperature approach (°C)</td>
<td>25.0</td>
</tr>
<tr>
<td>Heat exchangers minimum temperature approach (°C)</td>
<td>10.0</td>
</tr>
<tr>
<td>Recuperators minimum temperature approach (°C)</td>
<td>5.0</td>
</tr>
<tr>
<td>Heat exchanger pressure drop (%)</td>
<td>2.0</td>
</tr>
</tbody>
</table>
Figure 2: Scheme of the calcium looping process integrated with the recompression supercritical CO\textsubscript{2} cycle

3 PROCESS ANALYSIS

3.1 Considerations

Having adapted the recompression s-CO\textsubscript{2} cycle to recover the excess heat from the CaL plant, the retrofitted system needs to be benchmarked with the systems currently considered as state of the art. Therefore, the performance of the base configuration is compared with the CaL plant integrated with the supercritical reheated steam cycle. The comparison is made to the best configuration from Hanak et al. [45] in which oxycombustion of coal is used to provide heat for sorbent regeneration in the calciner. Moreover, in both configurations, the CaL plant is retrofitted to the 580 MW\textsubscript{el} supercritical coal-fired power plant to achieve the total CO\textsubscript{2} capture level of 90%.

A common feature of each Brayton cycle is the fact that the cycle efficiency can be further improved by introducing more reheat and intercooling stages, which would
increase the average temperature of heat addition and decrease the average temperature of heat rejection, respectively. Despite additional cost associated with the additional equipment, such designs would allow for more compact design of reheaters and intercoolers [44]. Dostal et al. [34] have estimated that the first reheat stage results in about 1.2% points improvement in the s-CO₂ cycle efficiency, while the second reheat stage adds only about 0.5% points to the efficiency. Their study also revealed that intercooling can increase the s-CO₂ cycle efficiency by up to 0.8% points. Although introducing more than one reheat and intercooling stage may not be an economically attractive option [34], it is worth exploring their effect on the net efficiency of the retrofitted system. Moreover, the fact that CO₂ is above its supercritical pressure (74 bar) throughout the cycle allows turning it to the dense phase by cooling it below the critical temperature (31.3°C). As a result, a pump can be used in place of the main compressor to increase the CO₂ pressure. Therefore, the following cases are considered in the configuration sensitivity study:

- Effect of reheat considering up to three expansion stages.
- Effect of intercooling by considering up to three compression stages.
- Effect of using a pump instead of the main compressor.

The performance of each thermodynamic cycle can be improved by increasing the operating temperature and pressure at the turbine inlet. Therefore, another sensitivity analysis is conducted to assess the effect of the high-pressure turbine inlet pressure and temperature on the net efficiency of the retrofitted system by varying the pressure between 200–300 bar and the temperature between 540–640°C. Finally, by combining both sensitivity studies the performance of the optimum design is estimated.
Table 5: Comparison of the key performance indicators

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Reference coal-fired power plant [25]</th>
<th>CaL retrofit (supercritical steam cycle) [45]</th>
<th>CaL retrofit (s-CO$_2$ cycle)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>System performance indicators</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gross power output (MW$_{el}$)</td>
<td>580.4</td>
<td>1023.3</td>
<td>1024.3</td>
</tr>
<tr>
<td>Total auxiliary electricity consumption (MW$_{el}$)</td>
<td>27.7</td>
<td>223.5</td>
<td>217.9</td>
</tr>
<tr>
<td>Net power output (MW$_{el}$)</td>
<td>552.7</td>
<td>799.9</td>
<td>806.4</td>
</tr>
<tr>
<td>Gross thermal efficiency (% HHV)</td>
<td>40.4</td>
<td>39.2</td>
<td>40.1</td>
</tr>
<tr>
<td>Net thermal efficiency (% HHV)</td>
<td>38.5</td>
<td>30.6</td>
<td>31.6</td>
</tr>
<tr>
<td>Net specific chemical energy consumption (kW$<em>{ch}$/kW$</em>{el}$)</td>
<td>2.6</td>
<td>3.3</td>
<td>3.2</td>
</tr>
<tr>
<td>Specific coal consumption (g/kW$_{el}$h)</td>
<td>350.3</td>
<td>438.2</td>
<td>425.4</td>
</tr>
<tr>
<td><strong>Integration impact indicators</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Increase in the net power output (%)</td>
<td>-</td>
<td>44.7</td>
<td>45.9</td>
</tr>
<tr>
<td>Net efficiency penalty (% HHV points)</td>
<td>-</td>
<td>7.9</td>
<td>6.9</td>
</tr>
<tr>
<td>Increase in net specific chemical energy consumption (%)</td>
<td>-</td>
<td>26.9</td>
<td>23.1</td>
</tr>
<tr>
<td>Carbonator CO$_2$ capture level (%)</td>
<td>-</td>
<td>79.6</td>
<td>80.0</td>
</tr>
<tr>
<td>Total CO$_2$ capture level (%)</td>
<td>0.0</td>
<td>90.0</td>
<td>90.0</td>
</tr>
<tr>
<td>CO$<em>2$ intensity factor (g/kW$</em>{el}$h)</td>
<td>792.3</td>
<td>111.6</td>
<td>108.7</td>
</tr>
</tbody>
</table>

### 3.2 Performance evaluation

Analysis of the retrofit of the 580 MW$_{el}$ coal-fired power plant with CaL plant using either the recompression s-CO$_2$ cycle or the supercritical steam cycle for excess heat recovery (Table 5) revealed that the gross and the net power output in both configurations were comparable and only slightly higher for the former cycle. Nevertheless, the net efficiency of the CaL integrated with the recompression s-CO$_2$ cycle was found to be 1%$_{HHV}$ point higher for the same high-pressure turbine inlet conditions. This is not only the result of a higher degree of high-grade heat utilisation in the recompression s-CO$_2$ cycle, but also due to more heat recovered to preheat the flue gas and the O$_2$ streams entering the CaL plant, which resulted in 2.9% reduction of the specific coal consumption of the retrofitted system (4.7% reduction of the coal
consumption in the calciner). As the specific capital cost ($/MW\textsubscript{el}) of the s-\text{CO}_2 cycle has been estimated to be up to 27% lower than that of the supercritical steam cycle operating in the same envelope \[34\], the recompression s-\text{CO}_2 cycle can be considered as a technically and economically viable option for decarbonisation of coal-fired power plants using CaL.

![Pie chart](image_url)

**Figure 3: Effect of the calcium looping secondary thermodynamic cycle selection on a parasitic load distribution for the 580 MW\textsubscript{el} coal-fired power plant retrofit scenario**

Analysing the distribution of the parasitic load (Figure 3), which again was estimated to be only slightly lower for the recompression s-\text{CO}_2 cycle, it is worth pointing out that the power requirement to run the ASU and the CCU compressors accounted for around 72.5% of the total system power requirement for both investigated configurations. As the remaining part of the parasitic load can be considered as unavoidable, further improvement of the retrofitted system’s performance can be achieved through a more efficient secondary cycle, higher level of heat integration in the CaL plant, alternative O\textsubscript{2} sources, such as chemical looping combustion \[71\] or
membranes [72,73] for oxy-combustion of fuel or heat sources to sustain sorbent regeneration and alternative CO₂ compression options.

3.3 Recompression supercritical CO₂ cycle sensitivity study

3.3.1 Hardware analysis

The impact of the number of reheating and intercooling stages, as well as the kind of pressure-change equipment (compressors or pumps) is presented in Figure 4. It can be clearly seen that change in the hardware has a small effect on the parasitic load distribution, as the reduction in the gross efficiency associated with the power requirements of the particular components of the retrofitted system varies by up to 0.1%HHV points. Hence, improvement of the retrofitted system’s performance is mostly associated with a higher cycle efficiency of the recompression s-CO₂ cycle.

The analysis revealed also that the net efficiency of the retrofitted system is more sensitive to the number of intercooling stages rather than to the number of reheating stages. Implementation of the first (T2C2) and the second (T3C2) reheat stage results in the net efficiency of the retrofitted system higher by 0.3%HHV points and 0.4%HHV points, respectively, with respect to the configuration without reheat (T1C2). Conversely, the net efficiency increase over the configuration without intercooling (T2C1) of 0.5%HHV points and 0.7%HHV points is reached on implementation of one (T2C2) and two (T2C3) intercooling stages, respectively. Hence, the base configuration with the single reheat and one intercooling stage appears to be an optimum one, as the addition of the second reheat and second intercooling stage results in net efficiency improvement of only 0.1%HHV points and 0.2%HHV points, respectively, and may not be economically viable. Importantly, the addition of intercooling allows reducing the clean flue gas temperature to 60°C, which is only
slightly above the flue gas discharge temperature in the reference coal-fired power plant (57.2°C), from 108.9°C. As a result, more low-grade heat is recovered from the CaL plant and used for power generation.

**Figure 4: Effect of recompression supercritical CO₂ cycle configuration on integrated process performance**

Additionally, implementation of one pump in place of the first compression stage in the base configuration (T2C1P1) resulted in a 0.4%\textit{HHV} point increase, while substitution of both compression stages with pumps (T2P2) resulted in only 0.1%\textit{HHV} point increase.

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\textsuperscript{a} TxCyPz indicates number of turbine stages $x$, compression stages $y$, and pumping stages $z$ in the configuration considered.

\textsuperscript{b} CaL s-SC refers to the reference configuration in which the excess high-grade heat from CaL is recovered in the supercritical steam cycle, while CaL s-CO₂ refers to the configurations in which the excess high-grade heat from CaL is recovered in the recompression s-CO₂ cycle.
in the net thermal efficiency. This is because CO$_2$ is in the dense phase at the pump discharge, and thus enters the LTR at a lower temperature. The former configuration results in the highest net thermal efficiency of the retrofitted system, which is 32.0%$_{HHV}$ corresponding to an efficiency penalty of 6.4%$_{HHV}$ points.

3.3.2 Turbine inlet conditions analysis

As shown in Figure 5, increase in the net thermal efficiency of the retrofitted system is directly proportional to the increases in both the turbine inlet temperature and pressure. Yet, the maximum turbine inlet temperature (640°C) was limited by the assumption that the reheater outlet temperature is equal to the high-pressure turbine inlet temperature and the minimum temperature approach in the reheater of 10°C. Nevertheless, Dostal et al. [34] claimed that the recompression s-CO$_2$ cycle operating at the maximum temperature of 650°C and pressure higher than 200 bar is the most promising layout for the s-CO$_2$ cycle. Although increasing the turbine inlet temperature to 640°C was found to increase the net thermal efficiency to 32.1%$_{HHV}$, which corresponds to 6.4%$_{HHV}$ points drop from the reference value, it would require a large heat exchange area to reach the desired minimum temperature approach. Hence, a turbine inlet temperature of 620°C, which resulted in net thermal efficiency of 31.9%$_{HHV}$ and is the maximum temperature for the most advanced supercritical steam cycles [44], would be more suitable for short- to mid-term applications. Moreover, a maximum turbine inlet pressure of 300 bar was selected, because such pressures are allowed by a very-high-grade steel, which is required to allow for high-temperature operation and to prevent the system from corrosion induced by the flue gas components [44]. It was estimated that the retrofitted system with the recompression s-CO$_2$ cycle operating with such turbine inlet pressure had the net thermal efficiency of 31.9%$_{HHV}$. 
Finally, it can be observed that the correlation with the temperature (Figure 5a) is nearly linear, while it is of second order for the pressure (Figure 5b). Such correlation for the latter parameter can be associated with the extent of the pressure drop in the heat exchangers, which increases with the cycle operating pressure. Analysis of the net thermal efficiency trends in Figure 5 indicated that further increase in the turbine inlet temperature, which would require a more complex heat exchanger network and
a multiple shaft turbine design, could result in an improvement of the process performance of about 0.1% per each 10°C increase in the turbine inlet temperature. On the contrary, increase in the turbine inlet pressure can be expected not to improve the net thermal efficiency of the retrofitted system much, mostly due to the associated increase in the pressure drop in the system.

3.3.3 Optimum recompression supercritical CO₂ cycle design

Having investigated how the performance of the retrofitted system varies with the recompression s-CO₂ cycle hardware and the turbine inlet conditions, the optimum configurations have been proposed and benchmarked against the base configuration in Table 6.

Table 6: Comparison of the key performance indicators for the optimised configurations

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Base case (T2C2)</th>
<th>Case 1 (T2C2)</th>
<th>Case 2 (T2C3)</th>
<th>Case 3 (T2C1P1)</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Turbine inlet conditions</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Temperature (°C)</td>
<td>593.3</td>
<td>620.0</td>
<td>620.0</td>
<td>620.0</td>
</tr>
<tr>
<td>Pressure (bar)</td>
<td>300.0</td>
<td>300.0</td>
<td>300.0</td>
<td>300.0</td>
</tr>
<tr>
<td><strong>System performance indicators</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Gross power output (MWₑ)</td>
<td>1024.3</td>
<td>1039.6</td>
<td>1045.7</td>
<td>1053.3</td>
</tr>
<tr>
<td>Net power output (MWₑ)</td>
<td>806.4</td>
<td>821.5</td>
<td>827.5</td>
<td>835.0</td>
</tr>
<tr>
<td>Gross thermal efficiency (%HHV)</td>
<td>40.1</td>
<td>40.8</td>
<td>41.0</td>
<td>41.3</td>
</tr>
<tr>
<td>Net thermal efficiency (%HHV)</td>
<td>31.6</td>
<td>32.2</td>
<td>32.4</td>
<td>32.7</td>
</tr>
<tr>
<td>Specific coal consumption (g/kWₑh)</td>
<td>425.4</td>
<td>416.9</td>
<td>413.9</td>
<td>410.1</td>
</tr>
<tr>
<td><strong>Integration impact indicators</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Net efficiency penalty (%HHV points)</td>
<td>6.9</td>
<td>6.3</td>
<td>6.1</td>
<td>5.8</td>
</tr>
<tr>
<td>CO₂ intensity factor (g/kWₑh)</td>
<td>108.7</td>
<td>106.5</td>
<td>105.7</td>
<td>104.7</td>
</tr>
</tbody>
</table>

It has been demonstrated in Section 3.3.2 that by increasing the turbine inlet temperature to 620°C, or increasing the pressure to 300 bar, the net thermal efficiency can increase by 0.3% HHV points. Unfortunately, no synergy effect occurred when both parameters were increased at the same time (Case 1), as the net thermal efficiency
identified in Table 6 increased by 0.6%\textsubscript{HHV} points over the base configuration, which is equal to the sum of increments incurred by a separate increase in the turbine inlet temperature and pressure. Moreover, allowing for higher capital cost, addition of the second intercooling stage (Case 2) improves the performance by another 0.2%\textsubscript{HHV} points, lowering the net efficiency penalty to 6.1%\textsubscript{HHV} points. Alternatively, by considering the pump in place of the first stage of the main compressor (Case 3), the retrofitted system performance can be improved by 0.5%\textsubscript{HHV} points, resulting in the net efficiency penalty of 5.8%\textsubscript{HHV} points. This corresponds to 1.1%\textsubscript{HHV} points improvement over the base configuration and 2.1%\textsubscript{HHV} points improvement over the best configuration for CaL with the supercritical steam cycle for coal-fired calciner reported by Hanak et al. [45].

4 CONCLUSIONS

This study proposed to use the recompression s-CO\textsubscript{2} cycle in place of the conventional steam cycle to recover high-grade heat from the CaL plant, which was retrofitted to the 580 MW\textsubscript{e} supercritical coal-fired power plant. The recompression s-CO\textsubscript{2} was adapted to allow for a high degree of heat integration and to utilise the low-grade heat. The net efficiency of the base configuration, which was characterised by single reheat and intercooling stages (T2C2), was estimated to be 1.0%\textsubscript{HHV} points higher compared to the reference supercritical steam cycle under the same turbine inlet conditions.

Further improvements in the net efficiency of the retrofitted system were achieved by varying the hardware and the turbine inlet conditions. It was found that adding the second reheat (T3C2) and second intercooling (T2C3) resulted in 0.1%\textsubscript{HHV} and 0.2%\textsubscript{HHV} points increase in the net efficiency of the retrofitted system, respectively. Also, replacing the first stage of the main compressor (T2C1P1) with the pump was found to increase the net efficiency by 0.4%\textsubscript{HHV} points. Furthermore, it was found that
increase of the turbine inlet temperature and pressure to 640°C and 300 bar would increase the net efficiency of the retrofitted system to 32.1%\textsubscript{HHV} and 31.9%\textsubscript{HHV}, respectively. However, considering the material availability and the short- to mid-term application of the proposed concept, the temperature of 620°C and pressure of 300 bar were selected for the optimal configurations. Depending on the hardware configuration, the net efficiency of the system can be increased to 32.2%\textsubscript{HHV} (T2C2), 32.4%\textsubscript{HHV} (T2C3) and 32.7%\textsubscript{HHV} (T2C1P1). The last configuration resulted in 2.1%\textsubscript{HHV} point net efficiency improvement over the reference CaL with the supercritical steam cycle (30.6%\textsubscript{HHV}). As the specific capital cost of the s-CO\textsubscript{2} cycle is up to 27% lower than that of the equivalent steam cycle, this study proves the viability of the recompression s-CO\textsubscript{2} cycle application in CaL. Since the steam cycles for a high-grade heat recovery and power generation are integrated with other high-temperature looping cycles, such as chemical looping combustion, it can be expected that these can also benefit from the s-CO\textsubscript{2} cycle implementation.

The analysis revealed also that the performance of the coal-fired power plant retrofitted with the CaL plant, in which the excess heat is recovered in the recompression s-CO\textsubscript{2} cycle, can be further improved by:

- higher level of heat integration in the CaL plant, alternative sources of O\textsubscript{2} for oxy-combustion of fuel or sources of heat for sorbent regeneration to minimise or avoid the power requirement associated with O\textsubscript{2} production;
- alternative processes and technologies to minimise the power requirement for CO\textsubscript{2} compression prior to transport;
- higher efficiency of the secondary cycle, which for the recompression s-CO\textsubscript{2} cycle can be achieved through alternative configurations allowing increasing the turbine inlet temperature.
NOMENCLATURE

\( a_1 \) Sorbent average conversion model fitting parameter  \\
\( a_2 \) Sorbent average conversion model fitting parameter  \\
b Sorbent average conversion model fitting parameter  \\
\( F_0 \) Fresh limestone make up rate kmol/s  \\
\( f_1 \) Sorbent average conversion model fitting parameter  \\
\( f_2 \) Sorbent average conversion model fitting parameter  \\
\( E_{CO_2} \) CO\(_2\) capture level in the carbonator -  \\
\( F_{CO_2} \) CO\(_2\) flow rate entering the carbonator kmol/s  \\
\( F_R \) CaO looping rate kmol/s  \\
\( f_{calc} \) Calcination reaction extent -  \\
\( f_{carb} \) Carbonation reaction extent -  \\
\( X_{ave} \) Average sorbent conversion -  \\
\( \Delta X_{carb} \) Carbonation conversion -

ABBREVIATIONS

ASU Air separation unit  \\
CaL Calcium looping  \\
CCS Carbon capture and storage  \\
CCU CO\(_2\) compression unit  \\
HRSG Heat-recovery steam generator  \\
HTR High-temperature recuperator  \\
LTR Low-temperature recuperator  \\
s-CO\(_2\) Supercritical CO\(_2\) cycle
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