Efficiency Improvement of Chemical Looping Combustion Combined Cycle Power Plants

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Chemical-looping combustion (CLC) is an innovative technology for power production with inherent carbon dioxide (CO₂) capture. Even though CLC imposes no direct energy penalty for CO₂ capture, previous works have shown significant energy penalties relative to natural gas (NG) combined cycle plants. This is due to the relatively low turbine inlet temperature (TIT), which is limited by the oxygen carrier used in the CLC process. Therefore, herein, an additional combustor (COMB) is included downstream of the CLC unit to raise the TIT (dependent on the CLC/COMB outlet temperature [COT] and the blade cooling). When NG is used in the additional COMB, the energy penalty is only 2.9% points with 72% CO₂ capture. Achieving higher CO₂ capture requires the use of H₂ fuel in the COMB. The efficiency of the H₂ production process plays an important role. For conventional H₂ production with post-combustion CO₂ capture, the added COMB brings no improvement and the energy penalty is 8.8% points. For an advanced H₂ production process (90% efficiency), the energy penalty reduces to 4.5% points with 100% CO₂ capture. The results show the potential of CLC-combined cycle power plants with an additional COMB to minimize the energy penalty of CO₂ capture.

1. Introduction

Rising atmospheric carbon dioxide (CO₂) concentrations are driving the scientific community to develop novel low-emission power production technologies. In addition, stringent emission policies are also being devised to mitigate CO₂ emissions.[1] Carbon capture and storage (CCS) is one strategy toward low-emission power production. Even though these strategies provide significant carbon capture capability, they are associated with significant energy penalty. Recent studies indicate that the traditional CO₂ capture technologies such as amine-based absorption integrated with natural gas (NG)-based power plants result in energy penalties of 7.6–8.4 %-points.[2–4]

Energy penalty is the primary cost driver of CO₂ capture. It increases fuel cost as well as capital cost due to increase in the plant size required to provide a certain power output. In addition, the costs associated with transport and storage increase the overall CCS costs significantly.[5] The higher fuel use is also associated with increased environmental impact, which counteracts some of the environmental benefits of CO₂ capture. Therefore, it is necessary to develop energy systems integrated with carbon capture facilities, which are highly efficient and low in energy penalty.

Chemical looping combustion (CLC) is an innovative process with inherent CO₂ capture at minimal energy penalty.[6] The energy penalty is only for compression to storage conditions due to segregated handling of the fuel and the oxidizer. An oxygen carrier (OC), which is generally a transition metal oxide,[7] is circulated between two interconnected reactors: a fuel reactor (FR), where the OC reduction by the fuel takes place producing CO₂ and steam (H₂O), and an air reactor (AR), where the OC is oxidized by the incoming air. The steam is condensed to obtain a pure stream of CO₂ ready for storage. The heat released in the AR maintains the thermal balance of the system and produces a high-temperature outlet stream that is used to drive a power cycle. Comprehensive details about the CLC process can be found in previous studies.[6,9] The generalized reactions in the two reactors are shown below.

Reduction 4MeO + CH₄ → 4Me + 2H₂O + CO₂ (1)
Oxidation 4Me + 2O₂ → 4MeO (2)

The power generation systems based on CLC are attractive due to the absence of the CO₂ separation step. However, the overall net electrical efficiency of CLC systems is limited by relatively low operating temperatures. The maximum operating temperature of the OC used in the CLC reactors is well below the inlet temperature that can be achieved by modern gas turbines (GTs). As outlined in the studies reviewed later, the resulting lower turbine inlet temperature (TIT) restricts the net electrical efficiency similar to that of a natural gas combined cycle (NGCC) plant integrated with CO₂ capture facility.

Several studies focusing on efficiency improvements in CLC with various power generation processes are reported in the

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Hassan et al. [14] performed a parametric study on a multistage improvement to the CLC plant proposed by researchers that the net electrical efficiency of these systems might reach higher than 60% with CO₂ generation of 0.33 kg-CO₂ kWh⁻¹. The efficiencies reported in these studies are promising; however, the studies were carried out considering simple power cycles with different strategies such as air saturation, which is generally not the case. These systems require a large amount of water, which is generally irrecoverable and will result in corrosion when the flue gases are cooled below dew temperature. Therefore, these systems are limited by the availability of water resources and increased risk of corrosion. Furthermore, the energy penalty for CO₂ compression to high pressures (HPs) was also excluded.

Naqvi et al. [12] introduced a steam cycle (SC) to recover the heat from the GT exhaust gases. A two-pressure heat recovery steam generator (HRSG) was used to produce the steam for power generation in steam turbines (STs). In addition, a three-stage-intercooled compression system for compressing CO₂ to pressures up to 200 bar was also included in the plant. They reported the net plant efficiency at full load to be 52.2%. This was a significant improvement to the CLC plant proposed by researchers before [6,10,11] considering the inclusion of a CO₂ compression system. Naqvi and Bolland [11] proposed a multipressure CLC plant with single- and double-reheat systems of the AR exhaust at the same oxidation temperature. They reported that the net electrical efficiency for a single-reheat system reaches above 53% at an oxidation temperature of 1200 °C. The double-reheat system resulted in a slight efficiency improvement over the single-reheat system. Hassan et al. [14] performed a parametric study on a multistage CLC plant by considering key operating parameters. It was reported that the overall net electrical efficiency could reach as high as 52% at stoichiometric conditions. Ekström et al. [15] reported the net thermal efficiencies for different CLC arrangements between 51% and 52%. Porrazzo et al. [16] developed a system with a detailed fluidized bed reactor model considering hydrodynamics and reaction kinetics. The net electrical efficiency obtained was 52.04%.

Zerobin and Pröll [17] developed a process model of a pressurized integrated CLC system and compared the performance with a simple gas turbine combined cycle (GTCC) plant consisting of a single-pressure HRSG. The objective of the study was to identify the limitations of HP CLC systems utilizing gaseous fuels. The net electrical efficiency reported for the GTCC plant with amine-based CO₂ capture was 49.43%, whereas that reported for the CLC system was 41.21% and 44.27% with 100% and 90% CO₂ capture, respectively. The low CLC efficiency is due to the conservatively low CLC operating temperature selected in the study (900 °C) and the 3 % -points CLC efficiency increase was achieved via additional firing after the CLC reactors to raise the TIT to 974 °C. The present study investigates the potential of additional firing in greater detail.

Petriz-Prieto et al. [18] investigated 15 different configurations consisting of three CLC systems, three power generation systems, and two OCs. The CLC systems considered were a simple two-reactor system, a three-reactor system with a calcination reactor for H₂ production (exCLC), and a three-reactor system with a steam reactor for H₂ production (CLC3). The power generation systems include an SC, a steam-injected gas turbine (STIG), and a humid air turbine (HAT) cycle. The OCs considered in this study were nickel based and iron based. For each of these cases, nine subcases were simulated by varying the reactor pressures (10, 20, and 30 bar) and TITs (1050, 1200, and 1350 °C). The average efficiencies reported for Ni-based OC plants: CLC with SC, CLC with STIG, and CLC with HAT were 45.92%, 47.4%, and 53.21%, respectively. Farooqui et al. [19] investigated the syngas production by H₂O/CO₂ splitting in a chemical looping unit and burning with oxygen from an air separation unit. The flue gas is expanded in a GT followed by heat recovery in an SC. The efficiency reported for the plant integration was 50.7%.

The aforementioned studies concerning the process simulations of a CLC gas-fired power plant are promising. However, the relatively complex dual circulating fluidized bed reactor that is the most widely used configuration for CLC systems presents technical challenges under pressurized operations. Despite these challenges, the pressurized operation of a dual reactor system has been demonstrated experimentally in the literature. Wang et al. [20] developed a laboratory scale unit of a CLC system using coke oven gas at a pressure of 3 bar. The continuous operation of the setup showed high fuel reactivity and resistance to OC agglomeration. Xiao et al. [21] at the Southeast University developed a 50 kWth dual circulating fluidized bed reactor system for solid fuels and demonstrated the operation up to a pressure of 5 bar. High carbon conversion with improved CO₂ purity and combustion efficiency was reported. A similar CLC unit of 50 kWth capacity utilizing coal as the fuel is under development at University of Kentucky, USA. [22] Korean Institute of Energy Research is developing a 0.5 MWth pressurized CLC system utilizing syngas, which could be operated at a pressure of 5 bar. [23] Further demonstration studies at higher pressures will be required to fully prove the feasibility of pressurized CLC in dual fluidized beds, but these studies suggest that it is technically feasible. In addition, there are several alternative chemical looping configurations under development to simplify pressurized operations such as packed beds, [24] gas switching reactors, [25] rotating reactors, [26] and internally circulating reactors. [27] These developments should accelerate the development of pressurized chemical looping technology.

The process parameters such as reactor temperature that governs the TIT, the pressure drop in the reactor, gas leakage between the two reactors, etc. have a significant influence on the net electrical efficiency. [17] Out of these parameters, the TIT is known to have a greater influence on net electrical efficiency. [28] As mentioned earlier, in typical CLC systems, the TIT is limited by the reactor temperature, which is maintained between 800 and 1200 °C. [29] This reactor temperature corresponds to the combustor outlet temperature (COT) in GTs. The TIT is defined at the first GT rotor and will be lower than the COT due to blade cooling. Increasing the COT beyond the aforementioned range is beneficial for the overall net electrical efficiency of a CLC plant. However, higher temperatures result in attrition and agglomeration of the OC material. Moreover, thermal sintering of the OC material occurs at about 70% of the melting point. [30] Hence, materials with a high melting temperature and high attrition and agglomeration resistance need to be developed. More research is being carried out on this aspect. [31] Despite this development, the reactor temperature...
(COT) could not be matched with that of the modern GTs, hence, the reduction in net electrical efficiency. One way to tackle this issue is to introduce a combustion chamber downstream of the AR. The fuel is burnt in the high-temperature oxygen-depleted air that raises the COT to the desired level.

Previous studies did not touch upon the significance of COT to the CLC plant efficiency much. The present study is motivated by recognizing this gap. Therefore, in the current study, a combustor (COMB) is added to the CLC combined cycle plant upstream of the GT to raise the COT beyond the achievable CLC temperature. The main objective of this work is to quantify the effect of varying the COT on the plant’s overall electrical and carbon capture efficiencies, using NG or hydrogen as fuel in the added COMB. As a secondary objective, the effect of the hydrogen production technology on overall plant performance will also be quantified. This understanding is critical to assess the potential of CLC in gas-fired power plants where GT technology is continuously developing toward a higher COT/TIT to maximize efficiency.

2. Methodology

2.1. Plant Configurations

System-level models of an NGCC plant and a CLC unit integrated with combined cycle configurations are developed. An additional COMB is included in the CLC plant to increase the COT. The impact of the two methods to mitigate possible NOx formation due to an increase in the COT is also investigated: steam injection (fuel dilution) and O2-depleted air recirculation. A description of these configurations is given in the following subsections.

The flow sheet for the reference NGCC plant is shown in Figure 1. The NGCC plant model is developed based on the recommendations in the European benchmark task force (EBTF) report.[32] Air is compressed and introduced into the COMB, whereas the fuel is preheated before injecting into the COMB. The high-temperature flue gases are expanded in a GT to near atmospheric pressure. The heat contained by the flue gas at the turbine outlet is further recovered in a SC. In the current study, the GE 9371FB GT system is considered which resulted in a COT of 1416 °C.[32] For simplicity, a single GT with a single HRSG and ST system is considered. The air compressor (AC), COMB, fuel preheater (PH), and the GT are simulated in Aspen plus, whereas the SC consisting of a triple-pressure single-reheat HRSG, STs, condenser (COND), and natural draft cooling tower (CT) is simulated in ThermoFlex. The HRSG comprised economizers, boilers, and superheaters that produce high-, intermediate-, and low-pressure (HP/IP/LP) steam. Subsequently, the steam is expanded in the respective STs for power generation. The plant specifications and the main assumptions used are listed in Table 1 and are referred from the EBTF report.[32]

Figure 2 shows the process flow diagram of CLC integrated with a combined cycle plant. NG is supplied to the FR where it undergoes an endothermic reduction reaction with the OC (NiO supported on NiAl2O4) producing CO2 and H2O. The reduced OC is then transferred to the AR. Compressed air is supplied to the AR where it undergoes an exothermic reaction with the incoming reduced OC. The heat released in the AR maintains the thermal balance by providing the heat required for the endothermic reaction in the FR and aids in the autothermal operation of the whole unit. The high-temperature exhaust from the AR, consisting of mostly N2, is expanded in the GT followed by heat recovery in a SC before releasing into the stack. Similarly, the FR exhaust is supplied to an expander (EX) for power generation and is used further for fuel preheating. The CO2-rich stream is sent to a three-stage compression unit where the steam is condensed and CO2 is compressed to a supercritical state for storage (P = 110 bar; T = 30 °C).

The operating conditions such as fuel and air mass flow rates and OC circulation rate of the CLC unit are kept similar to the base case plant of Naqvi and Bolland.[13] The amount of air used in the AR is 3.2 times the stoichiometric amount. This is to ensure the complete oxidation of the OC, which is about 1.3 times the stoichiometric amount required for the complete conversion of NG. Naqvi and Bolland[13] developed the CLC reactor

![Figure 1. Process flow sheet of a NGCC plant.](image-url)
Table 1. Plant specifications and main assumptions.

| Unit                  | Specification                                                                 |
|-----------------------|-------------------------------------------------------------------------------|
| NG [vol%]             | CH4—89%; C2H6—7%; C3H8—1%; C4H10—0.11%; CO2—2%; N2—0.89% (70 bar and 15 °C) |
| Air composition [vol%]| N2—77.3%; O2—20.7%; H2O—1%; Ar—0.92% (1.013 bar and 15 °C)                 |
| Hydrogen supply [C bar] | 15/14                                                                         |
| LHV/NG/H2 [kJ kg⁻¹]  | 46 502/119 800                                                                |
| Reactor/COMB pressure drop, [% of inlet pressure] | 5% | 92% |
| Air/H2 compressor polytropic efficiency | 92% |
| Gas/CO2 compressor polytropic efficiency | 85% |
| Compressors/turbines isentropic efficiency | 98% |
| Mechanical efficiency | 98% |
| SC (HRSG) ST system | Condensing reheat ST |
| Reheat temperature [°C] | Depends on each case |
| HP/IP/LP ST isentropic efficiency | Based on steam conditions and turbine size |
| HP/IP/LP steam pressure [bar] | 124/18.3/3.4 |
| HP/IP/LP steam temperature [°C] | Depends on each case |
| Pinch temperature/approach temperature [°C] | 15/5 |
| COND pressure [bar] | 0.048–0.071 (depends on each case) |
| Cooling system | Water cooling with natural draft CT |
| Water pump efficiency | 70% |
| Heat exchangers |                                                                 |
| Minimum temperature approach, gas–gas/gas–liquid [°C] | 10/10 |
| Pressure drop, % of inlet pressure | 1% |
| CO2 compression | 3 |
| Compression stages |                                                                 |
| Final CO2 condition [C bar⁻¹] | 30/110 |
| Compressor stages isentropic efficiency | 80/80/75% |
| CO2 pump efficiency | 75% |

heat and mass balance model by considering the reactor geometry, hydrodynamics, solids residence times, and transport between the two reactors. This resulted in the AR outlet temperature of 1200 °C. Details about the reactor model development can be found in the study by Naqvi et al. However, for simplicity in the current study, heat and mass balances are solved by considering the Gibbs energy minimization concept that resulted in the AR outlet temperature of 1150 °C for the same inlet specifications. This is a more conservative AR outlet temperature, given the limitations related to OC stability discussed earlier. Similar to the reference plant, the plant specifications and main assumptions are taken from the EBTB report and are listed in Table 1. The CLC unit including the AC, PH, GT, and CO2 EX are simulated in Aspen plus, whereas the SC is simulated in Thermoflex.

Figure 3 presents the CLC combined cycle plant with an additional COMB. The fuel supply, steam injection, and exhaust gas recirculation arrangements are also shown. The main object of adding a COMB is to increase the COT beyond the achievable CLC temperature. Different fuel flowrates (either NG or H2) are fed to the added COMB to vary the COT and evaluate its effect on the plant performance. Generally, a higher COT requires the greater use of EX blade cooling to mitigate the thermal stresses. The effect of changes in the amount of blade cooling on turbine efficiency is greatly dependent on the compressor pressure ratio and COT. The film cooling method is extensively used for blade cooling. In this method, compressor air is extracted and supplied into the blades. The amount of cooling air required is determined by using the following correlation based on the thermodynamic model given by Kim and Ro.

\[
\frac{m_c C_{p,c}}{m_b C_{p,g}} = C \frac{\varphi}{1 - \varphi}, \text{ where } \varphi = \frac{T_{gb} - T_b}{T_{gb} - T_c}
\]

where \(\varphi\) is the cooling effectiveness, \(T_g\) is the gas temperature flowing into the turbine, \(T_b\) is the coolant air temperature, \(T_c\) is the blade surface temperature, \(C\) indicates the level of blade cooling technology where a lower value corresponds to a high-level technology, \(m_c\) is the mass flowrate, and \(c_p\) is the specific heat.

The maximum blade material temperature allowed is usually between 800 and 900 °C. However, due to the application of thermal barrier coatings on turbine blades, the temperature could go higher. A constant blade temperature of 1000 °C has been assumed in the current study. The parameter \(C\) with a value of 0.04 for the first stage and increment of 10% for each stage is taken from the work of Kim and Ro. Four turbine blade stages have been considered in the current study. The pressure ratio across the stages is assumed to be constant. This allowed in identifying the temperature across the blade stages. The number of blade stages that requires cooling is then decided when this temperature goes below the assumed maximum temperature of the blade material. Using the blade cooling correlation, a single-stage blade cooling is required for the COT up to 1200 °C. For the COT up to 1400 and 1600 °C, blade cooling in two and three stages is required, respectively. Blade cooling is not considered for the CO2 EX as the maximum gas temperature is below the allowable blade material temperature.

The available GT module is used for both the NG and H2 fuel as it is robust to changes in fuel type and composition. The NG is split from the same supply line as the CLC unit and preheated using the FR exhaust before injecting into the COMB (not shown in the flow diagram). In contrast, H2 is compressed to the desired pressure (1.5 times the oxidizer) before injecting into the COMB. The source of H2 is considered from the study by Nazir et al. in which the supply line is at 15 °C and 14 bar, under the assumption that it is produced on site.

The AR exhaust is used as the oxidizer in the COMB. In this configuration, the air required in the AR is compressed to about 19.2 bar as opposed to 18.24 bar in conventional CLC shown in Figure 2. This is done to keep the flue gas pressure at the GT outlet similar to that in the CLC configuration. Consequently,
the AR outlet temperature obtained is 1160 °C as opposed to 1150 °C in the reference CLC plant. Considering the high-temperature exhaust and high flammability of H₂, steam injection and exhaust O₂-depleted air recirculation are included to reduce the flame temperatures in the COMB and limit NOx formation. The required IP steam is bypassed and compressed to the required pressure, before mixing with the H₂. In contrast, the O₂-depleted air is bypassed upstream of the stack and mixed.
with the incoming air. It is to be noted that the total air mass flow rate to the AR is kept constant.

2.2. Process Modeling and Performance Indicators

The heat and mass balance calculations were performed by considering the thermodynamic equilibrium in all the components. The Redlich–Kwong–Soave equation of state with Boston–Mathias alpha function (RKS-BM) is used to evaluate the properties. This property method is used as it is recommended for hydrocarbon processing applications. The CLC reactors and the COMB are modeled using RGIBBS module, which assumes chemical and phase equilibriums based on the Gibbs energy minimization concept. The separation of solid and gaseous streams is assumed perfect and modeled using a cyclone block with the incoming air. It is to be noted that the total air mass flow rate to the AR is kept constant.

For the SC, a condensing reheat ST system is considered. The model is developed such that the reheat temperature and the HP/IP/LP steam temperatures adjust automatically depending on each case. Furthermore, the ST efficiencies also adjust automatically based on steam conditions and turbine size required. Similarly, the COND pressure also varies depending on the steam condition. Water cooling with a natural draft CT system is adopted for compressor interstage and COND cooling.

For a consistent comparison of all the cases considered, the plant performance is evaluated using net electrical efficiency, efficiency penalty (%-points), and carbon capture efficiency (%) as shown in the following equations.

Net electrical efficiency:

$$\eta_{\text{net}} = \frac{W_{\text{turbines}} - W_{\text{compressors}} - W_{\text{pumps}} - W_{\text{auxiliary}}}{m_{\text{fuel}} \times \text{LHV}_{\text{fuel}}} \quad (4)$$

Efficiency penalty: $$\eta_{\text{pen}} = \eta_{\text{ref}} - \eta_{\text{plant}} \quad (5)$$

CO₂ capture efficiency: $$\eta_{\text{CO₂}} = \frac{\text{CO₂ captured}}{\text{CO₂ produced}} \quad (6)$$

3. Results and Discussions

Table 2 presents the stream information consisting of temperatures, pressures, and compositions at key locations in the NGCC plant. The COT obtained is 1416 °C, which is kept as the benchmark for comparison with other plant configurations. The net electrical efficiency obtained is 58.17% without CO₂ capture. For the CLC combined cycle plant, the steam information at key locations for the base case is given in Table 3. The COT obtained for the GT is 1150 °C and for the CO₂ EX is 835 °C. The net electrical efficiency obtained is 49.39% with 100% CO₂ capture. This configuration is considered as the base case for the CLC combined cycle power plant.

### 3.1. Effect of AR Outlet Temperature

For the CLC combined cycle reference case, the AR outlet temperature (COT) is varied by varying the amount of air into the AR. The air flow rate is varied from 697 to 1051 kg s⁻¹ (2.65–4 times the stoichiometric amount). The effect of this change on the net electrical efficiency and the energy penalty compared with the NGCC plant is shown in Figure 4. As expected, the net electrical efficiency increases significantly with increasing COT. As the COT increases from 1000 to 1300 °C, the net electrical efficiency increases from 44.94% to 52.46%. This increase in efficiency is due to the reduction in air compression work. It is evident from the results that the CLC power plant can have efficiencies better than the NGCC plant with conventional post-combustion CO₂ capture if the AR can be operated at 1200 °C or higher. However, it could be problematic to operate the reactor at such high temperatures due to OC degradation and limitations related to the materials used in the reactor body and downstream cyclone. Therefore, in the current study, an additional COMB is used to raise the COT from the base case of 1150 °C.
3.2. Plant Performance with an Additional COMB

The fuel used in the COMB is either NG or H2. As mentioned earlier, the supply conditions of H2 are taken from the chemical looping reforming (CLR) plant by Nazir and Bolland.[38] Hence, a compressor is also required to compress H2 to the desired pressure, which adds extra compression work. The COT considered for the additional COMB cases is 1416°C, which is the same as the NGCC plant.

Figure 5 depicts the net electrical efficiency, the carbon capture efficiency, and the energy efficiency penalty for the NGCC plant without CO2 capture, the CLC combined cycle plant, the CLC plant with NG-fired COMB, and the CLC plant with H2-fired COMB. There are various H2 production processes with different production and carbon capture efficiencies.[38,40–42] Therefore, the source of H2 from the conventional production process such as steam methane reforming with post-combustion CO2 capture to the most ideal H2 production processes is considered in the current study. For ease of comparison, four subcases are considered with different H2 production and carbon capture efficiencies. The H2 production efficiency is defined as the ratio between

| Table 3. Stream data of CLC combined cycle plant. |
|-----------------------------------------------|
| Point | T [°C] | P [bar] | Mass flow [kg s⁻¹] | CH4 | C2H6 | N2 | O2 | CO2 | H2O | Ar | NiO | Ni | NiAl2O4 |
|-------|--------|---------|---------------------|-----|------|----|----|-----|-----|----|-----|----|--------|
| 1     | 15     | 1.01    | 832.0               | 0   | 0    | 77.39 | 20.74 | 0.03 | 1.01 | 0.83 | 0   | 0   | 0   |
| 2     | 422    | 18.24   | 823.0               | 0   | 0    | 77.39 | 20.74 | 0.03 | 1.01 | 0.83 | 0   | 0   | 0   |
| 3     | 1150   | 17.53   | 767.5               | 0   | 0    | 82.40 | 15.61 | 0.03 | 1.08 | 0.88 | 0   | 0   | 0   |
| 4     | 497    | 1.04    | 776.6               | 0   | 0    | 82.40 | 15.61 | 0.03 | 1.08 | 0.88 | 0   | 0   | 0   |
| 5     | 108    | 1.01    | 776.6               | 0   | 0    | 82.40 | 15.61 | 0.03 | 1.08 | 0.88 | 0   | 0   | 0   |
| 6     | 10     | 70.00   | 15.0                | 89  | 8.11 | 0.89 | 0    | 2    | 0   | 0   | 0   | 0   | 0   |
| 7     | 434    | 18.44   | 15.0                | 89  | 8.11 | 0.89 | 0    | 2    | 0   | 0   | 0   | 0   | 0   |
| 8     | 835    | 16.63   | 70.5                | 0   | 0    | 0.28 | 34.66 | 65.06 | 0   | 0   | 0   | 0   |
| 9     | 466    | 1.05    | 70.5                | 0   | 0    | 0.28 | 34.66 | 65.06 | 0   | 0   | 0   | 0   |
| 10    | 30     | 110.00  | 40.0                | 0   | 0    | 0.81 | 98.97 | 0.21 | 0   | 0   | 0   | 0   |
| 11    | 37.5   | 1.26    | 95.4                | 0   | 0    | 0    | 0    | 0    | 100  | 0   | 0   | 0   |
| 12    | 37.4   | 0.06    | 95.3                | 0   | 0    | 0    | 0    | 0    | 100  | 0   | 0   | 0   |
| A     | 1150   | 17.53   | 616.0               | 0   | 0    | 0    | 0    | 0    | 0    | 78.01 | 0.00 | 21.99 |
| B     | 835    | 16.63   | 560.5               | 0   | 0    | 0    | 0    | 0    | 0    | 23.36 | 54.65 | 21.99 |

Figure 4. Effect of AR outlet temperature (COT).
H₂ LHV output and the equivalent NG LHV input to the reforming process. The equivalent NG LHV input also accounts for imports or exports of electricity and steam. Carbon capture efficiency is given in Equation (6).

1) Ideal case: 100% efficiency and 100% capture. This case assumes that zero-carbon hydrogen is available with no efficiency penalty. Since hydrogen must be produced by converting other energy carriers, this case is thermodynamically impossible. It is only presented as a benchmark to quantify the overall losses involved in the following real H₂ production scenarios. 2) A conventional method such as steam methane reforming with post-combustion CO₂ capture: 70% efficiency and 80% capture. 3) An advanced method such as membrane-assisted chemical looping reforming (MA-CLR): 80% efficiency and 90% capture. 4) An advanced method with a high degree of process integration where the steam in the CLC FR outlet stream is efficiently used in the reforming process: 90% efficiency and 100% capture. Thus, the thermal energy required for steam generation, which is the largest energy penalty in advanced reforming processes, is avoided. This could be achieved by either feeding part of the CLC FR outlet stream to an MA-CLR process or using a two-phase flow heat exchanger to recover the condensation enthalpy from the CLC FR outlet stream for generating steam for reforming.

The results show that, when compared to the NGCC plant, the energy penalty for a CLC base case is 8.8 %-points. This is similar to the expected penalty of an NGCC plant with post-combustion capture. When using an additional COMB fired by NG, the net electrical efficiency obtained is 55.31% with an energy penalty of only 2.9 %-points. However, since NG is used as the fuel in the COMB, there is a significant amount of CO₂ produced which is released into the atmosphere after heat recovery. The resultant carbon capture efficiency obtained for this case is about 72%. Another aspect that requires attention is the combustion phenomena with high-temperature O₂-depleted air. At such conditions, the combustion observed is slow and produces relatively large quantities of carbon monoxide (CO) and H₂ in the COMB. However, the high O₂-depleted air temperature at the COMB inlet should ensure complete fuel combustion. More research is required in this area to minimize pollutant formation. This is beyond the scope of the current study.

When H₂ is used as the COMB fuel, the ideal net electrical efficiency obtained is 55.31%, which is similar to that obtained by NG firing. In the NG case, the fuel required for the CLC cycle and the COMB is preheated to 439 °C. However, in the H₂-firing case, the NG required in the CLC cycle is preheated to 420 °C, which allowed for H₂ fuel preheating to 288 °C while maintaining the PH minimum temperature difference. Therefore, in the H₂-firing case, the thermal input required in the COMB is slightly higher to maintain the COT of 1416 °C (≈4.8 MW). Consequently, relative to NG firing case, the AC consumption is 0.08% less and GT production is 0.2% more. Furthermore, the power consumption of the H₂ compressor is 0.25% of the thermal input. The overall effect balances out in the net electrical efficiency. A carbon capture efficiency of 100% is obtained since there is no carbon involved in the COMB. The stream data at key plant locations are presented in Table 4 corresponding to Figure 3. It is to be noted that the power consumption and generation of different plant components are presented in terms of % of LHV of the total fuel input. In this way, it is easier to compare the performance of each component relative to the thermal input. As the fuel thermal input is different in different cases, using absolute values will give a wrong impression of the plant performance. The difference in fuel thermal input is due to the varying fuel consumption to maintain different COTs.

For the three different H₂ production methods mentioned earlier (conventional, advanced, and advanced with process...
integration), the net electrical efficiency ranges from 49.38% to 53.64% and the carbon capture efficiency from 93% to 100% (Figure 5, cases CLC_H2, b–d). The energy penalty ranges from 8.8 to 4.5 %-points. Thus, it is evident that using a H2-fired COMB to increase the COT can result in a CLC process that is more efficient than the NGCC plant with conventional CO2 capture. However, if conventional H2 production with CO2 capture is used, the added COMB does not provide a significant benefit.

### 3.3. Plant Performance with NG-Fired COMB

In this section, the performance of the CLC combined cycle plant with the NG-fired COMB is discussed. As mentioned earlier, the net electrical efficiency obtained is higher than that obtained in the NGCC plant with conventional CO2 capture. However, carbon capture efficiency is significantly reduced. The effects of varying the COT and AR outlet temperature are discussed in the subsequent sections.

#### 3.3.1. Effect of COT

The effect of varying COT on the plant net electrical efficiency and carbon capture efficiency is shown in Figure 6. The variation in COT is achieved by varying the amount of NG burnt in the COMB. The COT is varied from 1200 to 1600 °C which corresponds to 0.89–10.16 kg s⁻¹ of NG in the COMB. The largest COT is chosen since modern GTs such as the Mitsubishi J-series turbines can achieve a TIT of 1600 °C. The results show that as the COT is increased, the net electrical efficiency increases, whereas the carbon capture efficiency decreases significantly. The highest net electrical efficiency obtained is 57.63% with a COT of 1600 °C, which is close to the NGCC plant without CO2 capture (a COT of 1416 °C). The improvement in net electrical efficiency is due to the reduction in power consumption of the AC and increased power generation by the STs with respect to the thermal input. The corresponding carbon capture efficiency is only 59.25%, which is low compared to the base case CLC combined cycle power plant (100% capture).

#### 3.3.2. Effect of AR Outlet Temperature

Figure 7 shows the effect of varying the AR outlet temperature on the net electrical and carbon capture efficiencies. The temperature is varied from 1000 to 1300 °C by controlling the air flow rate in the AR (1111–721 kg s⁻¹). Moreover, the COT is kept constant at 1416 °C by controlling the NG flow rate into the COMB (12.39–2.13 kg s⁻¹). The results show that with an increase in the exhaust temperature the net electrical efficiency falls from 56.1% to 53.9%, whereas the carbon capture efficiency increases from 54.38% to 86.94%. The fall in the net electrical efficiency is due to the reduction in the mass flow rate of the AR exhaust. Consequently, the power generation in the GT and subsequently in the STs is also reduced. However, the power generation in the CO2 EX increases due to an increase in the FR temperature. Nevertheless, the net effect of combusting a larger fraction of the fuel in the CLC reactors on the net electrical efficiency is negative. The two main reasons as to why fuel combustion in CLC is less efficient than fuel combustion in the added COMB are that 1) energy recovery from the FR outlet is less efficient due to the lower temperature of that stream and 2) more CO2 compression.
is required. The rise in the carbon capture efficiency is due to the reduction in the NG flow rate into the COMB.

3.4. Plant Performance with a Hydrogen-Fired COMB

In this section, the performance of the CLC combined cycle plant with H₂-fired COMB is discussed. Using H₂ as the fuel in COMB minimizes the CO₂ emissions since only the emissions during H₂ production need to be considered. The effect of COT on plant performance with the four different performance levels of the H₂ production process considered in Figure 5 will be discussed in this section.

Furthermore, studies on methods for lowering the flame temperature in the COMB to reduce NOx formation are also presented. The actual design of the COMB to be used in the proposed plant is beyond the scope of this study. However, it can be pointed out that the high COMB inlet temperature will

Figure 6. Effect of COT.

Figure 7. Effect of AR outlet temperature.
eliminate any constraints related to igniting and sustaining a flame because the fuel will spontaneously combust in this hot O₂-rich environment. On the other hand, the high COMB inlet temperature will require special materials to maintain the structural integrity of the COMB and could, therefore, impose another constraint on the maximum allowable CLC AR temperature.

Regarding NOx formation, it is noted that the COMB will achieve ideal lean pre-mixed combustion with almost no NOx formation in either of two limits: perfectly distributed fuel injection (an infinite number of infinitely small fuel injectors) or perfect mixing (an infinite amount of turbulence). Naturally, neither of these limits can be achieved in practice, but the design criteria of the COMB to achieve minimal NOx are clear: maximize the number of fuel injectors and maximize the amount of turbulence (swirl). In case NOx cannot be controlled sufficiently via COMB design, the effects of two further NOx reduction mechanisms on plant performance are investigated in this section: fuel dilution and air recirculation.

### 3.4.1. Effect of COT

Figure 8 shows the effect of varying the COT on plant performance. The COT is varied from 1200 to 1600 °C by controlling the H₂ flow rate into the COMB (0.34–4 kg s⁻¹). The results are presented for four subcases considered. In Figure 8a, it is shown that the net electrical efficiency increases in the range 50.37–57.68% with increasing COT. This is because of the reduction in power consumption of AC and an increase in ST power generation with respect to the fuel thermal input. Figure 8b shows the plant performance when a conventional H₂ production process is considered. An insignificant variation in net electrical efficiency with COT is observed. The highest net electrical efficiency of 49.46% is obtained at a COT of 1400 °C. On further increasing the COT to 1600 °C, a slight drop in the efficiency is observed. On the other hand, the carbon capture efficiency decreases from 98.44% to 90% with increasing COT. This is because of the increase in the H₂ fuel input to the COMB, which increases the equivalent NG that is used during the H₂ production process. This reinforces the conclusion drawn from Figure 5 that the use of H₂ in the added COMB will not be useful if conventional steam methane reforming with post-combustion CO₂ capture is used to supply H₂.

Figure 8c shows the results when an advanced H₂ production process is considered. The net electrical efficiency increases in the range from 49.69% to 52.35% with increasing COT. On the other hand, carbon capture efficiency decreases from 99.31% to 95.36%. Figure 8d shows the results for an advanced H₂ production process with a high level of process integration. Similar to other cases, net electrical efficiency increases from
50.07% to 55.19% when COT is increased. There is no change in carbon capture efficiency as 100% carbon capture is assumed for this H₂ production process. A highly efficient H₂ production can, therefore, result in the attractive performance of a CLC combined cycle plant with an added COMB. Future work will investigate process integration schemes to achieve the performance shown in Figure 8d.

3.4.2. Effect of Steam Dilution

Steam dilution is considered as a measure to lower the flame temperature of H₂ combustion, thereby suppressing thermal NOₓ formation. The required steam is extracted after the HP ST and compressed to 27.36 bar (1.5 times the oxidizer pressure). The steam dilution is varied from 0% to 50% by volume and mixed with H₂ before entering into the COMB. In this case, COT is maintained at 1500 °C, which is achieved by controlling the H₂ flow rate in the range of 3.03–3.78 kg s⁻¹. The results obtained for the net electrical efficiency and the carbon capture efficiency are shown in Figure 9.

It is observed that more steam dilution requires more H₂ to maintain the COT, thus increasing the power consumption of the H₂ compressor. Increasing steam dilution also increases power consumption by the steam compressor. In addition, there will be less steam available for power production. Relative to the thermal input, there is a 2.41% reduction in power produced by STs when steam dilution is increased from 0% to 50%. Furthermore, there is also a decrease in AC consumption by 2.68%. As a result, the net electrical efficiency decreases. A similar trend is observed in all the H₂ production cases considered.

Figure 9a shows the results for an ideal H₂ source. There is a slight drop in net electrical efficiency from 56.67% to 56.25% with 100% carbon capture efficiency. Figure 9b shows the results when a conventional H₂ production method is considered. The net electrical efficiency drops from 49.42% to 48.13%, whereas the drop in carbon capture efficiency is from 91.43% to 90.33%. Figure 9c shows the results when the advanced H₂ production process is considered. The highest thermal and carbon capture efficiencies obtained are 52.2% and 95.5%, respectively. The corresponding drops in these efficiencies with 50% steam dilution are 1% and 0.5%, respectively. Figure 9d shows the results for the advanced H₂ production case. The net electrical efficiency decreases from 54.59% to 53.89% with 100% carbon capture.

The results indicate that the influence of steam dilution on the net electrical efficiency is small. It increases the mass content of the exhaust gas and maintains power generation from the GT to be around 70% of the fuel thermal input in all the cases. This is a positive finding in case future studies show that high quantities

![Figure 9](https://www.advancedsciencenews.com/)

**Figure 9.** Effect of steam dilution for the cases with added H₂ firing for different H₂ and carbon capture efficiencies: a) 100%, 100%; b) 70%, 80%; c) 80%, 90%; and d) 90%, 100%.
of steam dilution are necessary to suppress pollutant formation in the added COMB.

3.4.3. Effect of O₂-Depleted Exhaust Gas Recirculation

The idea behind the recirculation of O₂-depleted exhaust gas is to reduce the O₂ content of the stream entering into the COMB. This will result in O₂-poor local regions close to the flame, limiting the flame temperature and NOx formation. The exhaust gas is recirculated from a point just before the stack and varied from 0% to 50% by mass. The exhaust gas temperature at the stack is around 80 °C, which affects the AC exit temperature. This, in turn, increases the AR exhaust temperature. Hence, the air flow rate is varied from 872 to 937 kg s⁻¹ to maintain the AR exhaust temperature at 1160 °C. The COT is maintained at 1500 °C by controlling the H₂ flow rate into the COMB.

As shown in Figure 10, the net electrical efficiency is not much affected by exhaust gas recirculation. Due to an increase in air flow rate, the power consumption of the AC increases by 6.7%, whereas the power generation from the gas and STs increases by 5.1% and 1.6%, respectively. The net effect of this variation is negligible over the range of exhaust gas recirculation considered. For the ideal H₂ production case, the average net electrical and carbon capture efficiencies obtained are 56.62% and 100%, respectively. For the conventional H₂ production case, the net electrical and carbon capture efficiencies obtained are around 49.11% and 91.11%, respectively. For the advanced H₂ production case, the efficiencies obtained are around 51.98% and 95.88%, respectively. Finally, for the advanced case with improved process integration, the efficiencies obtained are around 54.46% and 100%, respectively.

Exhaust gas recirculation has an even smaller effect on process efficiency than steam dilution. Therefore, any amount of dilution could be used without compromising the process efficiency. However, a more detailed analysis of the COMB is required to find the optimum value of exhaust gas dilution for the combustion phenomenon to occur smoothly.

3.4.4. Effect of CLC Cycle Pressure

The effect of varying the CLC cycle pressure on plant performance is shown in Figure 11. Three cycle pressures are investigated: 19, 30, and 50 bar. For the 30 and 50 bar cases, two- (PR = 5.47) and three-stage (PR = 3.69)-intercooled air compression systems are used, respectively. The heat removed during intercooling is not utilized and simply discarded. It is to be noted that the AC exit temperature is limited to 500 °C due to compressor material limitations. The COT, in this case, is kept

![Figure 10. Effect of exhaust gas recirculation for the cases with extra H₂ firing for different H₂ and carbon capture efficiencies: a) 100%, 100%; b) 70%, 80%; c) 80%, 90%; and d) 90%, 100%.](image-url)
constant at 1416 °C by maintaining the H2 flow rates into the COMB at 2.26, 2.49, and 2.5 kg s⁻¹, respectively. Similarly, the AR outlet temperature is also maintained constant at 1160 °C by keeping the air flow rates at 861, 942, and 940 kg s⁻¹, respectively. The H2 is compressed to 1.5 times the oxidizer pressure in the COMB. No steam dilution and exhaust gas recirculation are considered in the current simulations.

For the ideal H2 production case, the net electrical efficiency decreases with increasing pressure. The drop in efficiency is 5.47 %-points with an increase in pressure from 19 to 50 bar. This is due to the increase in AC work from 39.65% to 59.77%, increase in GT power generation from 73.91% to 95.78%, and a decrease in the ST power generation from 19.22% to 11.42% of the fuel thermal input. For the conventional H2 production case, the net electrical efficiency decreases by 1.01 %-points when the pressure is increased to 30 bar and then further decreases by 4.21 %-points when the pressure is increased to 50 bar. The carbon capture efficiency obtained also follows a similar trend and is averaged at 92.5%. For the advanced H2 production case, the highest efficiency of 51.69% is obtained at 19 bar and it drops by 5.33 %-points at 50 bar. The average carbon capture efficiency for this case is 96.56%. For the advanced H2 production case with improved process integration, the drop in net electrical efficiency is 5.41 %-points when the pressure is increased from 19 to 50 bar, with a small decrease in efficiency by 0.2 %-points. For comparison, the same pressure increase in the case with a COT of 1416 °C caused a 0.81 %-point reduction in efficiency. When the pressure is further increased to 50 bar, the efficiency drops by 3.53%, as opposed to 4.6 %-points, in the case with COT of 1416 °C.

While it may be possible to improve the efficiency of the higher pressure cases slightly by integrating the heat removed in intercooling into the SC, these results seem to suggest that the optimum process pressure is at the lower end of the investigated range with a slight increase in the optimal pressure with increasing COT.

3.5. Comparison of Energy Penalty for Carbon Capture

The energy penalty for carbon capture in an NGCC power plant with post-combustion capture ranges from 7.6 to 8.4 %-points.[2–4] This includes the energy required for separation of CO2 from the flue gas and the compression to a supercritical state. The separation step consumes a significant amount of energy, which is avoided in CLC systems. In CLC systems, the only direct energy penalty aside from a small pressure drop is for CO2 compression. Despite this, the net electrical efficiency for a typical CLC plant obtained in this study is only 49.39% with an energy penalty of about 8.78 %-points when compared to the NGCC plant without carbon capture due to the limitation in the maximum CLC operating temperature.

When this limitation is overcome using an additional COMB after the CLC unit, the energy penalty can be as low as 4.53 %-points when the fuel source is H2 from an advanced H2 production process. If NG is used in the added COMB, the energy penalty reduces to 2.9 %-points at the expense of a lower carbon capture efficiency (72%). The added COMB can allow the CLC plant with extra firing to benefit from continued advances in GT technology, leading to gradual efficiency gains over the coming decades.

Figure 11. Effect of CLC cycle pressure at TIT = 1416 °C for different H2 and carbon capture efficiencies: a) 100%, 100%; b) 70%, 80%; c) 80%, 90%; and d) 90%, 100%.
Other strategies for increasing the efficiency of CLC systems reported in the literature include multistage CLC systems\(^{[13,14]}\) and integration with HAT power cycles.\(^{[18]}\) Naqvi and Bolland\(^{[13]}\) and Hassan et al.\(^{[14]}\) studied multistage CLC systems operated at different cycle pressures and reported the energy penalty of 5.17 and 6.17 %-points, respectively, when compared to our NGCC plant (relatively inefficient reference plants were reported in these studies). The disadvantage of these systems is the significant increase in the capital costs and plant complexity. Petriz-Prieto et al.\(^{[18]}\) calculated impressively high efficiencies of CLC configurations integrated with a HAT cycle, but this advanced power cycle can also be applied to a reference plant without CO2 capture and holds no inherent advantages with respect to improving the efficiency of CLC.

However, the exCLC\(^{[47]}\) and CLC3\(^{[48]}\) configurations investigated by Petriz-Prieto et al.\(^{[18]}\) produce H2 which can be combusted to reach a high TIT. It was shown that these CLC configurations returned about 1–3 %-points lower efficiency than conventional CLC for the same TIT.\(^{[18]}\) From the data at a COT of 1300 °C in Figure 4 and 8 in this study, it can be seen that this efficiency reduction corresponds roughly to added firing with H2 produced at an efficiency of 80% (the efficiency at COT = 1300 °C in Figure 8c is 1.53 %-points lower than the same COT in Figure 4). It should also be mentioned that these three-reactor CLC systems\(^{[47,48]}\) will be more complex to scale-up and operate and also require more advanced OCs than a conventional two-reactor CLC.

**4. Conclusions**

A system-level model of a CLC combined cycle power plant incorporating an additional COMB after the CLC unit is developed. The objective of adding a COMB is to increase the TIT by increasing the COT by firing either NG or hydrogen. The reactor temperatures in CLC are limited by the OC material as well as the materials used in the construction of the reactor, the downstream cyclone, and the additional COMB.

When it is assumed that the CLC outlet temperature is limited to 1150 °C, the efficiency of the resulting plant is only 49.4% compared with 58.2% for the NGCC benchmark—an energy penalty of 8.8 %-points. Additional NG firing after the CLC unit to increase the COT to the same level as the benchmark plant (1416 °C) reduced the energy penalty to only 2.9 %-points while achieving 72% CO2 capture. Such a plant with moderate CO2 capture at a very low energy penalty could be economically attractive in an environment with moderate CO2 prices. When CO2 prices eventually become high, this plant could be retrofitted to fire the additional COMB with H2 instead of NG, thereby avoiding the CO2 emissions from combusting additional fuel after the CLC reactors.

For the use of H2 in the additional COMB, several cases were investigated with different H2 production efficiencies. If the H2 production efficiency is only 70% (representative of current steam methane reforming with post-combustion CO2 capture), the additional COMB brings no significant benefit. However, for a highly efficient H2 production process with integrated CO2 capture with an efficiency of 90%, the overall plant energy penalty reduces to only 4.5 %-points with 100% CO2 capture. Such efficiencies could be achieved by the efficient process integration of H2 production technologies based on chemical looping reforming that are currently under development.

The CLC plant with an additional COMB can also benefit from continued improvements in GT technology to allow for a higher COT. In the case with a 90% H2 production efficiency, a COT of 1600 °C produced a further efficiency gain of 2.98 %-points.

Although the design of the additional COMB was out of the scope of this study, the investigation of the effect of two NOx

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**Figure 12.** Effect of CLC pressure at TIT = 1600 °C for efficiencies: H2 = 90%; CO2 = 100%.
control mechanisms was also completed: steam dilution and exhaust gas recirculation. The negative effects on plant efficiency were small to negligible, implying that NOx can be controlled without serious effects on plant performance.

Overall, the results show that adding a COMB and increasing the TTT significantly increase the net electrical efficiency and enable future gains from advanced GTs allowing for very high CO2. Additional firing with NG or H2 can be adapted according to CO2, pricing trends in the future, whereas potential NOx formation challenges can be circumvented with minimal additional energy penalties. Therefore, it can be concluded that a CLC combined cycle plant with an additional COMB has the potential to become a commercial technology and merits further research.

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Conflict of Interest
The authors declare no conflict of interest.

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