Development of multimode gas-fired combined-cycle chemical-looping combustion-based power plant layouts

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Abstract
Operation of power plants with carbon dioxide capture and sequestration (CCS) and without carbon dioxide capture and storage modes and energy penalty or energy utilization in such operations is of great significance. This work reports on two gas-fired pressurized chemical-looping combustion (CLC) power plant layouts with two inbuilt modes of flue gas exit, namely, one with carbon dioxide capture mode and the second mode is letting flue gas (consists of carbon dioxide and water) without capturing carbon dioxide. Without CCS mode, the higher thermal efficiencies of 54.06 and 52.63% are obtained for natural gas and syngas, respectively. In carbon capture mode, a net thermal efficiency of 52.13% is obtained with natural gas and 48.78% with syngas. The operating pressure of the air reactor is taken to be 13 bar for realistic operational considerations, and that of the fuel reactor is 11.5 bar. Two power plant layouts were developed based on combined-cycle chemical-looping combustion (CC CLC) for natural gas and syngas fuels. A single layout is developed for two fuels with a possible retrofit for dual fuel operation. The CLC power plants can be operated with two modes of flue gas exit options, and these operational options make them higher thermal efficient power plants.

Keywords Chemical looping combustion · Layout · Gaseous fuels · Energy balance · Multimode · Carbon capture

Notation
- CLC: Chemical-looping combustion
- CC: Combined cycle
- CC CLC: Combined-cycle chemical-looping combustion
- CCS: Carbon capture and sequestration (or storage)
- HRSG: Heat recovery steam generator
- IGCC: Integrated gasification combined cycle
- NGCC: Natural gas combined cycle
- SMOC: Steam moderated oxyfuel combustion
- \( h_{\text{Air}} \): Enthalpy in air entering into air reactor, kJ/kg
- \( h_{\text{DepAir}} \): Enthalpy of oxygen depleted air reactor leaving the system, kJ/kg
- \( h_{\text{MeO}} \): Enthalpy in oxygenated carrier, kJ/kg
- \( h_{\text{Me}^{-1}} \): Enthalpy in reduced oxygen carrier, kJ/kg
- \( h_{\text{Fuelin}} \): Enthalpy flow in fuel entering into fuel reactor, kJ/kg
- \( h_{\text{Exhaust}} \): Enthalpy flow in exhaust leaving fuel reactor, kJ/kg
- \( \Delta H_{\text{Ox}} \): Heat of oxidation, kJ/mol
- \( \Delta H_{\text{Red}} \): Heat of reduction, kJ/mol
- \( M_{\text{Exhaust}} \): Mass flow of exhaust (containing CO\(_2\) and water vapor) leaving fuel reactor per unit time, kg/s
- \( M_{\text{O}_2,\text{in}} \): Mass of oxygen in fresh air, entering to air reactor, kg/s
- \( M_{\text{DepAir}} \): Mass of depleted air at the exit of air reactor, kg/s
- \( M_{\text{O}_2,\text{out}} \): Mass of oxygen present in depleated air at the exit of the air reactor, kg/s
- \( M_{\text{Fuelin}} \): Mass of fuel entering into fuel reactor, kg/s
- \( M_{\text{Exhaust}} \): Mass of exhaust gas (containing CO\(_2\) and water vapor) leaving fuel reactor, kg/s
- \( M_{\text{MeO}} \): Mass of oxidized oxygen carrier, kg/s
- \( M_{\text{MeO}^{-1}} \): Mass of reduced oxygen carrier, kg/s
- \( P_1 \): Compressor inlet pressure, bar
- \( P_2 \): Compressor outlet pressure, bar
- \( r_p \): Compression pressure ratio
- \( T \): Temperature at any point, °C
- \( T_1 \): Temperature of the gas at compressor inlet, K
- \( T_2 \): Isentropic temperature of gas at compressor exit, K

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Introduction

The emission of greenhouse gas, primarily carbon dioxide from power stations is a major concern for energy sectors, climate change, and terrestrial ecosystems (IPCC 2014, 2021). At present, carbon dioxide emission from power sectors alone is 35–40% of the total CO2 emission, and in these sectors, fossil fuels account for the anthropogenic CO2 emissions. The global average atmospheric carbon dioxide concentration in the year 1850 was 278 ppm, and this was increased in the last decade with an average growth of 2.4 ppm/year, and in the year 2020 atmospheric concentration of CO2 is 416 ppm (IEA 2016, 2021). This shows the atmospheric concentration of CO2 has increased globally by about 50% between the years 1850 to 2020. There are many ways to reduce CO2 gas emissions from the industrial sector, including combined heat and power (CHP), mineral adsorption, energy efficiency, fuel switching, use of renewable energy, carbon capture utilization and storage (CCUS) measures, and use of fuel cells powered by natural gas or renewable hydrogen (IPCC 2021; IEA 2021; Masson-Delmotte et al. 2021). Most combustion technologies involve energy penalties in flue gas cleaning and subsequent carbon capture. This should be minimized to implement CCS with the least energy penalty. There are several advantages with gas fuels (for example, natural gas and syngas) over high carbon emissive solid fuels to make power generation less CO2 intensive. The dependency on coal-based power generation will continue in countries like India, China, and South Africa for the next 40 years (Jayanti et al. 2012).

Emissions of carbon dioxide from power plants and industries have been of great interest by means of carbon capture and sequestration (IPCC 2014). Three principal ways to capture CO2 from flue gas upon combustion in power plants are post-combustion capture, pre-combustion capture, and oxy-fuel combustion (Ghonieum 2011). Comparative analysis of natural gas-fired oxyfuel and CLC-based power plant layouts is made by Basavaraja and Jayanti (2015a) for atmospheric and combined-cycle (CC) modes and net efficiency reported for oxyfuel-based CO2 recycle and combined-cycle steam moderated oxyfuel combustion (CC SMOC) is, respectively, 30.93 and 46.57% and CLC atmospheric and combined-cycle chemical-looping combustion (CC CLC) is, respectively, 43.11 and 51.94% after incorporating energy penalty for CO2 compression to 110 bar. Though state of art has been illustrated in these technologies with technical maturity (Wall 2007), many of these involve energy penalties in CO2 capture. Energy penalty for coal-based, pre-combustion capture, and compression are between 7.5 and 10% (Finkenrath 2011; Vasudevan et al. 2016) post-combustion capture and compression is between 8 and 12% (Goto et al. 2013; Wang et al. 2017, 2018; Song et al. 2019) and for oxyfuel combustion capture is between 9.5 and 12% (Jayanti et al. 2012; Jenni et al. 2013; Navajas et al. 2019). The energy penalty integrated gasification combined-cycle (IGCC) shell varies between 9.4 and 10.5%, and natural gas combined-cycle (NGCC) post-combustion capture is between 4.7 and 5.8% (Davison and Thambimuthu 2009; Zhao et al. 2021).

The concept of chemical-looping combustion was proposed in 1954 to produce pure CO2, and later research work to increase thermal efficiency on the reversibility of combustion processes using the chemical-looping concept is made (Lewis and Gilliland 1954; Richter and Knoche 1983; Ishida et al. 1987; Ishida and Jin 1994). The important parameters of CLC-based plants, such as gas–solid contact pattern, oxygen carrier reactivity at different temperatures for various fuels, and temperature distribution in a reactor system, are now well understood by research (Adanez et al. 2012; Tijani et al. 2017; Alam et al. 2020, Hu 2021). Studies on CLC-based power plants show that energy loss will be minimal (Naqvi et al. 2007; Kvamsdal et al. 2007; Navajas et al. 2019; Surywanshi et al. 2021; Wang et al. 2021) in oxygen separation from the air and such studies made in integrated gasification combined-cycle (IGCC) power plant with and without CO2 capture (Petrescu and Cormos 2017). Jin and Ishida (2001), Adanez et al. (2006a), Abad et al. (2007), Qasim et al. (2021), and Barros do Nascimento et al. (2022) studied on synthesis and kinetics of oxygen carriers used in pressurized CLC environments in lab scale. These results cannot be used in the scale-up reactor, but these show kinetics in 1 to 15 bar range CLC system will be feasible in case of heavy-duty CLC reactors. Ooi and his co-workers (2014) studied retrofitting power plants for carbon sequestration and problems associated with carbon-constrained energy planning (CCEP).

Operation of power plant with CCS involves a number of factors like fuel type, electricity cost, and geographical area for sequestration and environmental issues (Tola and Pettinaiu 2014). Detailed analysis on fuel switching for CLC-based power plant layout was made by Basavaraja and Jayanti (2015b) to address alternative fuel as an energy source for atmospheric CLC plants by thermodynamic, transport, and kinetic factors considerations. Studies on pressurized/combined-cycle-based CLC system operation (Wolf 2004; Naqvi et al. 2007; Zang 2018) have been reported in the literature.
These literature studies show operating CLC-based units with high thermal efficiency without much energy consumption for air separation with the feasibility of handling flue gas in with CCS and without CCS modes. Pressurized CLC shows higher thermal efficiency, and there has been the possibility of operating in both with CCS and without CCS modes. In the literature, detailed combined-cycle CLC-based power plant layouts with an option to handle flue gas with CO$_2$ capture and free let-out and fuel flexibility have not been properly addressed. Against this background, the purpose of the present work is to make a detailed single power plant layout incorporating carbon capture and without carbon capture options and ready to accommodate gaseous fuels of different compositions. In the present scenario in which carbon capture and sequestration (CCS) is being considered seriously for implementation within the lifetime of soon-to-be-built power plants and the prevailing opinion is in the favor of gas-fired power plants.

In this work, pressurized CLC reactors-based power plant layouts development studies made from the detailed mass and heat balances, power cycle calculations, and thermodynamic analysis for both natural gas and syngas fuels. This paper focuses on the development of a single layout of a gas-fired combined-cycle power plant based on CLC that operates with both syngas and natural gas fuels with carbon capture (ready to accommodate CCS as and when it becomes mandatory) or without carbon capture.

The objective of this work is to develop a gas-fired power plant layout incorporating two ways of handling flue gas with a prospective to know heat generation sections and heat utilization to generate electrical power using Brayton and Rankine cycles while still ensuring options of handling flue gas containing CO$_2$ with CCS and without CCS routes.

This paper aims to study, develop a process flow sheet and analyze two gas-fired pressurized chemical-looping combustion power plants and their configuration for two modes of operations, i.e., one with carbon capture mode and the other one is allowing flue gas from the plant to the atmosphere thereby enabling retrofitting possibility for fuel switch.

Three power plants based on pressurized chemical-looping combustion are considered and are:

(a) natural gas-fueled plant with and without carbon capture modes
(b) a syngas-fueled plant with and without carbon capture modes, an
(c) a future-ready plant operating with and without carbon capture modes with a retrofit for an option to fuel switch.

Detailed paper structure is shown in Fig 1 in the form of a block diagram.

**Depiction of mass and energy calculation**

**Pressurized chemical-looping combustion**

The principle of pressurized chemical-looping combustion is shown in Fig. 2. Here, the fuel combustion is split into two stages. Firstly, a solid metal (in a low oxidation state, denoted as MeO$_x$-1) is oxidized by oxygen in the air to form metal oxide (completely oxidized state, denoted as MeO$_x$) in an air reactor. In the later stage, the metal oxide gives oxygen to react with the hydrocarbon fuel during fluidization to form carbon dioxide and water vapor in the fuel reactor. Two-stage combustion of fuel by reactor systems eliminates oxygen separation units and avoids the mixing of N$_2$ with flue gas. The exit gases from the CLC reactor system are taken through gas turbines followed by heat recovery steam generators to generate power and CO$_2$ separated from the flue gas upon cooling sent for compression. Pressurized operation of CLC reactor systems enables combined power cycles operated under gas turbine as per Brayton cycle and steam turbine as per Rankine cycle to get improved net thermal efficiency of power plant (Kehlhofer 1999; El-Wakil 2010).

In recent years, pressurized CLC is gaining importance from the research in the view of reactor configuration, solid–gas contact, reaction kinetics, fuel conversion (Abad et al. 2007; Xiao et al. 2010, 2012; Zheng et al. 2014). Studies made by Adanez et al. (2006) and Garcia-Labiano et al. (2006) show that reduction of NiO to Ni oxygen carrier conversion remains 100% for CH$_4$, H$_2$, CO, and O$_2$, and conversion is reduced at higher pressure. In the pilot plant studies (Erlach et al. 2011) showed no attrition and no agglomeration when that Ni/NiO was operated at 1300 °C in CLC units.

The heat required for the endothermic metal oxide reduction (by fuel) is supplied from the oxidized metal coming from the air reactor. Reaction in two CLC reactors is by gas–solid contact during fluidization. Based on the reactive nature of oxygen carrier with air and fuel, the air reactor operates in circulating fluidized bed mode, and the fuel reactor operates in bubbling fluidized bed mode. Due to varying types of fluidization in air and fuel reactors results in more pressure drop and to account for this pressure in the fuel reactor is assumed to be 11.5 bar (at 1100 to 1150 °C) with the solid conversion of 80%. A similar condition (Naqvi 2006; Adanez et al. 2006) was used in literature for pressurized CLC systems. This operating pressure is chosen in the present study for realistic operation of CLC reactor system. The gases expanded to one bar from gas (air and exhaust) turbine are further cooled by heat recovery steam generators to run steam turbines of Rankine cycle and water.
Introduction on CO₂ emission from power sectors and literature review on oxy-fuel, pre combustion and post combustion capture technologies, oxygen carriers, chemical-looping combustion based power plants and their operation with CCS. Identification of literature gap in pressurized CLC based power plant operated in fuel switching (incorporating with and without carbon capture modes)

Fig. 1 Structure of the paper

- General description of pressurized chemical-looping combustion based power plant operates with CO₂ capture and without CO₂ capture modes
- Energy balance model for CLC reactors
- Development of natural gas fired pressurized CLC power plant layout based on heat balance data and thermodynamic analysis
- Development of syngas fired pressurized CLC power plant layout based on heat balance data and thermodynamic analysis
- Power cycle model for pressurized chemical looping combustion
- Development of fuel flexible pressurized CLC plant lay-out that has with CCS and without CCS modes operating options (based on heat balance and thermodynamic analysis of natural gas and syngas fired power plant layouts)

In this study, pressurized CLC power plant air reactor operating temperature is taken to be 1200 °C (at 13 bar operating pressure) for both natural gas and syngas fuels, and the operating temperature for pressurized CLC power plant fuel reactor is taken to be 1150 °C (at 11.5 bar operating pressure) and 1226 °C (at operating pressure 11.5 bar), respectively, for natural gas and syngas (Adanez et al. 2006; Naqvi et al. 2007; Erlach et al. 2011; Zhang et al. 2018; Rajabi et al. 2019; Joshi et al. 2021). The isentropic efficiency of the gas turbine, compressors efficiency, and water pump efficiency is taken to be, respectively, 92%, 85%, and 75% (Wolf 2004; Naqvi et al. 2007; Basavaraja and Jayanti 2015a; Khan et al. 2020). Steam cycle operating pressures for syngas pressurized CLC power plant are taken to be 150 bar (at 450 °C), 20 bar (at 325 °C), and 1.7 bar (at 190 °C) as inlet pressures to the high-pressure, medium-pressure, and low-pressure turbines, respectively (Basavaraja and Jayanti 2015a; Wolf 2004). Steam cycle operating pressures for natural gas pressurized CLC power plant are taken to be 1.7 bar (at 190 °C) as inlet pressures at low-pressure turbine (El-Wakil, 2010). The energy balance and combined-cycle arrangement are discussed in “Energy balance” section.
Energy balance

In this section, mass and energy balance details for the combined-cycle CLC system are given. The Nickel oxide (NiO) supported on Nickel di-aluminum tetroxide (NiAl₂O₄) in a 60:40 weight ratio is chosen as oxygen carrier (Brandvoll and Bolland 2004; Naqvi et al. 2007; Erlach et al. 2011; Lucia Blas et al. 2016; Zhang et al. 2018; Rajabi et al. 2019). The nickel oxide flow rate calculation is made by considering 25% excess oxygen supply to fuel in the fuel reactor (Basavaraja and Jayanti 2015a). The CLC-reactors are assumed to be adiabatic with isothermal and homogenous mixing of solids with gases (Naqvi et al. 2007; Rajabi et al. 2019). Airflow rate is calculation is considered in an air
reactor with an ~220% of excess air (Wolf 2004; Naqvi et al. 2007; Rajabi et al. 2019). Considering major components of the natural gas and syngas fuels, and their reaction with NiO is given as per reactions (1), (2), and (3), respectively with excess oxygen (Adanez et al. 2012):

\[ \text{CH}_4 + 4\text{NiO} \rightarrow 4\text{Ni} + 2\text{H}_2\text{O} + \text{CO}_2 \quad \Delta H = 133.6 \text{kJ/mol CH}_4 \]  

(1)

\[ \text{CO} + \text{NiO} \rightarrow \text{Ni} + \text{CO}_2 \quad \Delta H = -48 \text{kJ/mol CO} \]  

(2)

\[ \text{H}_2 + \text{NiO} \rightarrow \text{Ni} + \text{H}_2\text{O} \quad \Delta H = -15 \text{kJ/mol H}_2 \]  

(3)

Nickel is oxidized to nickel oxide in air reactor as per reaction (4) given below

\[ 2\text{Ni} + \text{O}_2 \rightarrow 2\text{NiO} \quad \Delta H = -468 \text{kJ/mol O}_2 \]  

(4)

In reactions (1) to (4), values of heat of reaction shown have been estimated from the values available in the literature for the temperature range 1150 to 1230 °C (Adanez et al. 2012; Linderotholm et al. 2008). Based on reactions (1) to (4), stoichiometric oxygen requirement for syngas is calculated, and in similar way, other components of natural gas are estimated. One can see here that syngas composition consist of major portions of carbon monoxide, hydrogen, and lessor part of methane. The reaction of CO and H2 with NiO is exothermic compared to lessor CH₄ gas endothermic reaction with NiO. This makes the overall syngas reaction with NiO mildly exothermic.

In this system, power is generated primarily by the Brayton cycle and bottoming Rankine cycle. The basis for all CLC calculations is the amount of fuel converted in the reduction/fuel reactor. In combined-cycle CLC, a metal oxidation reaction is taken up by the large mass of excess air. With the given fuel flow, the amount of oxygen converted is found from reaction stoichiometry, and the heat balance for the air reactor can be solved. Air and fuel reactors heat balance (as per Fig. 4) is formulated respectively according to Eqs. (5) and (6):

\[
M_{\text{Air in}} h_{\text{Air in}} + M_{\text{MeO}_x} h_{\text{MeO}_x} - (M_{\text{O}_2\text{in}} - M_{\text{O}_2\text{out}}) \Delta H_{\text{Ox}} = M_{\text{Dep Air}} h_{\text{Dep Air}} + M_{\text{MeO}} h_{\text{MeO}}
\]  

(5)

\[
M_{\text{Fuel in}} h_{\text{Fuel in}} + M_{\text{MeO}_x} h_{\text{MeO}_x} - M_{\text{Fuel in}} \Delta H_{\text{Red}} = M_{\text{Exhaust}} h_{\text{Exhaust}} + M_{\text{MeO}_x} h_{\text{MeO}_x}
\]  

(6)

*Fig. 4* Schematic of energy flow in combined-cycle chemical-looping combustion
In Eq. (5), terms $h_{\text{MeO}}$, $h_{\text{Me} \times -1}$, $h_{\text{Airin}}$, and $h_{\text{DepAir}}$ are, respectively, the enthalpy in oxidized metal, reduced metal, air entering, and oxygen-depleted air leaving the air reactor. In Eq. (6), terms $h_{\text{Fuelin}}$ and $h_{\text{Exhaust}}$ are respectively, the enthalpy of fuel entering into fuel reactor and enthalpy of the exhaust (stream containing CO$_2$ and water vapor). $\Delta H_{\text{Ox}}$ is the heat of oxidation in kJ/mol i.e., heat released during the reaction of Ni with O$_2$ to form NiO. The term $\Delta H_{\text{Red}}$ is heat of reduction in kJ/mol i.e., the heat absorbed during the natural gas oxidation or NiO reduction.

Ni oxidation is an exothermic reaction and therefore the term $\Delta H_{\text{Ox}}$ associated with molar mass is with plus sign as given in Eq. (5) and NiO reduction in fuel reactor is endothermic for the natural gas-fired case and thus the term $\Delta H_{\text{Red}}$ associated with molar mass is with minus sign as given in Eq. (6) and same will be become plus sign for exothermic NiO reaction with syngas.

Here $M_{\text{O}_2}$, $M_{\text{DepAir}}$, $M_{\text{O}_2}$, $M_{\text{Fuelin}}$, and $M_{\text{Exhaust}}$ are, respectively, mass of oxygen in the fresh air, the mass of the depleted air, unreacted or excess oxygen leaving air reactor, the mass of fuel entering into fuel reactor, and mass of exhaust gas (containing CO$_2$ and water vapor) leaving fuel reactor, respectively. In Eqs. (5) and (6), $M_{\text{MeO}}$ and $M_{\text{MeO} \times -1}$ denotes oxidized and reduced states of the oxygen carrier, respectively.

Thermal input for natural and syngas fired combined-cycle CLC-based power plant is considered 761 and 800 MW$_{\text{th}}$, respectively, supplied in continuous fuel flow. The composition in mole % of natural gas (Linderholm et al. 2009) is taken as 89.51% CH$_4$, 5.92% C$_2$H$_6$, 2.36% C$_3$H$_8$, 0.40% isobutene, 0.56% n-butane, 0.13% isopentane, 0.08% n-pentane, 0.06% C$_6$H$_{12}$, 0.28% N$_2$, and 0.70 CO$_2$ with lower heating value of 49.17 MJ/kg. Syngas (that generates almost two times the quantity of CO$_2$ of that of natural gas upon complete combustion) molar composition (Winslow 1977) is taken to be 45.7% H$_2$, 19.6% CO, 6.6% CH$_4$, and 28.1% CO$_2$ with the lower heating value of 11.2 MJ/kg. In both fuel-fired cases, comprehensive mass and energy balance is made, and thermodynamic analysis is carried out on the gas preheaters, air reactor, the fuel reactor, gas turbines (depleted air and exhaust gas turbines) of Brayton power cycle, and steam turbines of Rankine power cycles. The combined power cycle for pressurized chemical looping combustion is described below.

Pressurized chemical-looping combustion power cycle

A schematic diagram of the pressurized CLC power cycle is shown in Fig. 5. The CLC Brayton cycle consists of an air compressor, depleted air turbine, and exhaust gas (CO$_2$ + H$_2$O) turbine. The fuel is assumed to be pressurized. The oxygen-depleted air from the air reactor and exhaust stream containing CO$_2$ and water vapor from the fuel reactor are drawn into two separate Brayton cycles and are expanded in an air turbine and

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Fig. 5 Arrangement of combined-cycle CLC power cycle
CO₂ turbine. The CLC reactor exit gases after expansion preheat the respective reactor inlet gases using gas–gas heat exchangers or recuperators. The heat left in the exhaust and depleted gas streams is extracted by using a series of heat exchangers or heat recovery steam generators (HRSG) to generate steam as per the Rankine cycle. Figure 5 shows a combined-cycle chemical-looping combustion power cycle arrangement having two Brayton cycles and a Rankine cycle.

**Results and discussion**

**Layout of natural gas-fired pressurized CLC**

In this section, detailed mass and energy balance for power plant and complete process flow sheet of pressurized CLC plant is discussed and details of these are given below.

**Mass and energy balances**

Mass and heat balances are made on a CLC reactor system containing five major CLC sub-units. These are the fuel oxidation reactor (operating at 1150 °C and 11.6 bar), the air oxidation reactor (operating at 1200 °C and 13 bar), the fuel and the air pre-heaters and coolant airflow (Adanez et al. 2006; Naqvi et al. 2007; Erlach et al. 2011; Zhang et al. 2018; Rajabi et al. 2019). Based on reactions (1) to (4), stoichiometric oxygen requirement for components of syngas and natural gas is calculated. Mass flow of the individual stream of these sub-units has been found by balancing moles of reactant and product based on the fuel component mole fractions (Basavaraj and Jayanti 2015). Using Eqs. (5) and (6) heat (energy) balances have been made for the natural gas-fueled combined-cycle CLC system of 761 MWₘₑ fuel input. Gas streams enthalpy is estimated using the temperature-dependent correlations for the heat capacities of the individual components (Nayef and Redhoune 2010), while those for the NiO (oxygen carrier) and NiAl₂O₄ (support) can be obtained using correlations given by Knacke et al. (1991) and Barin (1989), respectively. The resulting energy and mass flow rates in and out of these sub-units are shown in Fig. 6.

Compressed air (at 177 °C and 13.5 bar) at the mass flow of 870.12 kg/s is divided into two streams namely, coolant airflow of 58.44 kg/s and the process air of 811.68 kg/s. The air of mass flow 811.68 kg/s (with 215.55% excess

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**Fig. 6** Furnace side heat balance of 761 MWₘₑ natural gas-fueled combined-cycle chemical-looping combustion plant

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O₂) is drawn to the CLC reactor system, and heated air (at 420 °C) from the recuperator enters the air reactor where 59.7 kg/s of oxygen present in the air is selectively reacted with metal (containing 218.95 kg/s of Ni, 69.66 kg/s of NiO, and 232.21 kg/s of NiAl₂O₄) to form metal oxide stream (containing 348.31 kg/s of NiO and 232.21 kg/s of NiAl₂O₄). Conversion of NiO to Ni is 80% in the fuel reactor. The Energy produced by exothermic metal oxidation reaction in the air is taken out by N₂ rich air (of flow 751.99 kg/s) and metal oxide streams. The heat absorbed by the endothermic metal oxide reduction reaction in the fuel reactor is 113 MW. This is supplied by the oxygen carrier. 15.47 kg/s of natural gas is preheated from 30 to 350 °C by the exhaust gas of 75.1 kg/s. The O₂-depleted air from the air reactor is 751.99 kg/s at 1200 °C. The oxygen-depleted air and coolant air (58.44 kg/s at 177 °C) mix at the turbine inlet to form a total mass flow of 810.43 kg/s and a temperature of 1132 °C at the turbine inlet. The depleted air and coolant bleed mixture gases expand in the gas turbine. The depleted air turbine exit gas temperature at 1 bar is 497 °C. Exhaust gas temperature reduces to 666 °C upon expansion. The computed value γ (= Cp/Cv) for oxygen-depleted air and fuel reactor exhaust given are 1.345 and 1.226, respectively. These values are comparable with this reputed literature (Naqvi et al. 2007; Wolf 2004) for the same inlet gas composition for depleted air and exhaust turbines. Depleted air at the exit of the air reactor (at 1200 °C) mixes with the coolant air, so that turbine inlet temperature is taken to be 1132 °C (Adanez et al. 2006; Naqvi et al. 2007; Erlach et al. 2011; Zhang et al. 2018; Rajabi et al. 2019). Nearly atmospheric pressure depleted air and exhaust gas streams (after expansion in gas turbines) at low pressure are used for power generation (as per ranking cycle) after preheating respective reactor inlet gases. The complete process description for the combined-cycle chemical-looping combustion power plant flow diagram is given below (“Natural gas-fired power plant layout” section).

### Natural gas-fired power plant layout

The chemical-looping combustion-based combined cycle is unique compared to a conventional combined cycle as the depleted air from the air reactor and flue gas containing CO₂ and water vapor from the fuel reactor are drawn into two separate Brayton cycles and are expanded in an air turbine and CO₂ turbine. Based on the heat balance data (as shown in Fig. 6) and thermodynamic analysis (as shown in Table 1), combined-cycle natural gas-fired power plant layout is made. The combined-cycle chemical-looping combustion power cycle consists of four major components, namely compressor, recuperators, gas turbine, and steam turbine.

Since combustion is divided into oxidation and reduction reactions in two pressurized fluidized bed reactors, therefore two gas turbines are used to expand them in the proposed layout. Gases are compressed to desired reactors operation using compressors. Recuperators provide the necessary heat to preheat the reactants. The description of the streams in the process plant, as shown in Fig. 7, is described below.

### Table 1: Detailed thermodynamic analysis of natural gas-fueled 761 MW₉₈ combined-cycle chemical-looping combustion power plant

| Stream | T (°C) | P (bar) | m (kg/s) | h (kJ/kg) |
|--------|-------|--------|----------|-----------|
| **Air** |       |        |          |           |
| A1     | 30    | 303.15 | 1.01     | 870.12    | 0.00      |
| A2     | 152   | 425.15 | 3.43     | 870.12    | 123.71    |
| A3     | 35    | 308.15 | 13.50    | 870.12    | 5.02      |
| A4     | 177   | 450.15 | 13.50    | 811.68    | 149.37    |
| A5     | 177   | 450.15 | 13.50    | 58.44     | 404.77    |
| A6     | 420   | 693.15 | 13.50    | 811.68    | 404.77    |
| A7     | 1200  | 1473.15| 13.00    | 751.99    | 1298.12   |
| A8     | 497   | 770.15 | 1.01     | 810.43    | 491.62    |
| A9     | 241   | 514.15 | 1.01     | 810.43    | 216.94    |
| A10    | 234   | 507.15 | 1.01     | 810.43    | 209.92    |
| A11    | 74    | 347.15 | 1.01     | 810.43    | 44.34     |
| **Fuel** |      |        |          |           |
| F1     | 30    | 303.15 | 13.00    | 15.47     | 0.00      |
| F2     | 350   | 623.15 | 13.00    | 15.47     | 1607.00   |
| **Exhaust** |     |        |          |           |
| E1     | 1150  | 1423.15| 11.60    | 75.1      | 1720.79   |
| E2     | 1036  | 1309.15| 1.01     | 75.1      | 211.06    |
| E3     | 463   | 736.15 | 1.01     | 75.1      | 568.26    |
| E4     | 128   | 401.15 | 1.01     | 75.1      | 72.03     |
| E5     | 40    | 313.15 | 1.01     | 75.1      | 23.56     |
| E6     | 40    | 313.15 | 1.05     | 34        | 41.72     |
| E7     | 40    | 313.15 | 1.05     | 41.1      | 8.54      |
| E8     | 35    | 308.15 | 110.00   | 41.1      | 4.26      |
| **Oxygen carrier** | | | | |
| M1     | 1200  | 1473.15| 13.00    | 580.52    | 1254.49   |
| M2     | 1150  | 1423.15| 11.60    | 520.83    | 963.85    |
| **Steam/water** | | | | |
| S1     | 31    | 304.15 | 1.70     | 104.81    | 4.17      |
| S2     | 31    | 304.15 | 1.70     | 18.13     | 4.17      |
| S3     | 190   | 463.15 | 1.70     | 13.07     | 2852.17   |
| S4     | 31    | 304.15 | 1.70     | 86.68     | 4.17      |
| S5     | 31    | 304.15 | 1.70     | 46.95     | 4.17      |
| S6     | 190   | 463.15 | 1.70     | 46.95     | 2852.17   |
| S7     | 31    | 304.15 | 1.70     | 39.73     | 4.17      |
| S8     | 120   | 393.15 | 1.70     | 39.73     | 2709.16   |
| S9     | 190   | 463.15 | 1.70     | 39.73     | 2852.17   |
| S10    | 190   | 463.15 | 1.70     | 86.68     | 2852.17   |
| S11    | 190   | 463.15 | 1.70     | 104.81    | 2852.17   |
| S12    | 39    | 312.15 | 0.07     | 104.81    | 2571.70   |
| S13    | 30    | 303.15 | 1.01     | 104.81    | 0.00      |
The terms A1, A2, and A3 denote fresh air streams and are, respectively, the air before compression, air after the first stage of compression, and air at the exit of the second compression stage. The major part of the compressed air (A4) is first preheated and then admitted to the air reactor to oxidize the reduced metal oxide (M2). The depleted air (A7) mixes with the cooling air (A5) drawn from the air compressor and then expands down. The depleted air from the turbine exit (A8) preheats the fresh air (A4) to (A6). The depleted air from the turbine exit is further cooled from A9 to A10 by a water stream (S8) to generate steam (S9). Pressurized fuel (F1) after preheating (F2) reacts with the oxygenated metal carrier (M1) to form CO2 and H2O vapor mixture exhaust (E1). The expanded exhaust (E2) from the turbine preheats the fuel by cooling down to (E3) and further cools down (E4) by giving heat to water stream S2, which forms steam S3. The depleted air stream finally cools to 75 °C (A10 to A11) while heating water stream (S5) to stream (S6). Thus, the exit gases from the air turbine and exhaust gas are drawn into the Rankine cycle through a heat.

Fig. 7 Schematic of the natural gas-fueled combined-cycle chemical-looping combustion power plant without the provision for carbon capture
recovery steam generator to generate subcritical steam at 190°C (1.7 bar). The exhaust gas stream at 128°C is let into the atmosphere through a chimney when the power plant operates without CO₂ mode. To operate a power plant in carbon capture mode as shown in Fig. 8, the exhaust stream from the fuel reactor is admitted to flue gas conditioner for further cooling to ~40°C, and the condensed water vapor is separated (E6) and CO₂ gas (E7) is compressed to 110 bar.
(E8) at 35 °C. The isentropic efficiency of gas and steam turbines are assumed to be 92% (Wolf 2004; Basavaraja and Jayanti 2015a; Khan et al. 2020). The thermodynamic analysis of the natural gas-fueled CC CLC is shown in Table 1. The efficiency of the water pump and compressor are taken as 75 and 85%, respectively (Wolf 2004; Naqvi et al. 2007; Basavaraja and Jayanti 2015a; Khan et al. 2020).

**Layout of syngas fired pressurized CLC**

The mass and energy balance model is the same as already presented in the “Mass and energy balances” section for pressurized CLC with natural gas as the fuel has been used to determine the furnace side parameters for syngas firing. Since syngas has a higher cost of compression of CO2, the layout has been designed for 800 MWth, which amounts to 71.43 kg/s of syngas flow and is 4.6 higher than the flow rate of natural gas required for a 761 MWg plant. On the furnace side, the principal sub-systems considered are the air reactor, and its preheater, the fuel reactor and its preheater, the gas turbines used to extract power from these exhausts, and the compressor to supply air to the air reactors as well as coolant air to the air turbine. Syngas is assumed to be available under pressurized conditions, and the cost of its compression is not included in the analysis. As in the case of syngas, the air reactor is fixed to operate at 13 bar and 1200 °C, and the fuel reactor is fixed to operate at the pressure of 11.6 bar (at 1226 °C) (Adanez et al. 2006; Naqvi et al. 2007; Erlach et al. 2011; Zhang et al. 2018; Rajabi et al. 2019). The same oxygen carrier is used, and its flow rate has been determined for syngas requirements for an 800 MWth power plant. The operating temperature of the fuel reactor and the inlet and exit temperatures of the recuperators have been adjusted to account for the exothermicity of the reduction reaction in the syngas-fired fuel reactor. The specific heat capacity ratio (γ) is taken as 1.20 (Wolf 2004; Naqvi et al. 2007). The resulting mass and energy flow rates into these subsystems are shown in Fig. 9.

Based on the heat balance data (as shown in Fig. 9) and thermodynamic analysis (as shown in Table 2) combined-cycle syngas fired power plant layout is made. The process flow diagram for the CC CLC is shown in Fig. 10. The description of the streams in the process plant is briefly explained below. The inlet and exit streams reactor system is the same as the natural gas layout. The energy utilization from exit gas of the syngas-fired power plant is given below. The expanded exhaust (E2) from the gas turbine gets cooled down to E3 in the processing preheating the fuel and further to E4 and E5 by giving heat to water stream S2 to form high-pressure steam S3, which is superheated to S4 by the
cooling exhaust gas (E5 to E6). After expansion in the high-pressure turbine (S5), the exit steam is reheated (S6) and fed to the medium pressure (20 bar) steam turbine. After expansion to 1.7 bar in the medium pressure turbine, it is mixed with steam generated in various sections (S3, S6, and S16) of the process the plant from O₂-depleted air and exhaust gas steams via heat recovery steam generators to produce superheated steam at 1.7 bar at a temperature 190 °C. It then finally expands from 190 to 39 °C (at 0.069 bar). The exhaust gas (E6) at a temperature of 128 °C leaves the plant through the chimney to the atmosphere. The thermodynamic properties of the streams at various locations are shown in the thermodynamic analysis of the CC CLC is shown in Table 2.

### Schematic Diagram of the without CCS and with CCS enabled modes power plant

A schematic diagram of the proposed power cycle is shown in Fig. 5. It is based on pressurized chemical-looping combustion of gaseous hydrocarbon fuels such as natural gas, syngas, biogas, and shale gas. The combustion of hydrocarbon fuels occurs in two stages in two separate reactors. In the fuel reactor, the fuel combusts with a metal oxide, e.g., NiO supported on NiAl₂O₄ in a 60:40 weight ratio (taken in present work) prepared in the form of micron-sized particles, and gets converted to CO₂ and H₂O. In the process, the metal oxide gets reduced. It is therefore fed to the air reactor where it gets regenerated by reacting directly with air. Both the metal oxidation and the metal reduction reactions are carried in the range of 1150 to 1250 °C, depending on the reaction and on the fuel. A circulating fluidized bed type of configuration is envisaged for the cyclic oxidation/reduction reactions of the metal/metal oxide. It is also proposed that the reactions are carried out at a pressure of 13 bar in the air reactor and at a slightly reduced pressure of 12 bar in the fuel reactor. The high-temperature reactions in the reactors thus give rise to high-temperature gaseous products at a pressure of 12 to 13 bar. These are expanded in separate air or gas turbines to near atmospheric pressure. The expanded gases will still be hot having a temperature of 500 to 700 °C depending on the reactor and the fuel.

### Table 2 Detailed thermodynamic analysis of syngas-fueled 800 MWth combined-cycle chemical-looping combustion

| Stream   | T (°C) | T (K) | P (bar) | m (kg/s) | h (kJ/kg) | Stream   | T (°C) | T (K) | P (bar) | m (kg/s) | h (kJ/kg) |
|----------|--------|-------|---------|----------|-----------|----------|--------|-------|---------|----------|-----------|
| Air      |        |       |         |          |           | Oxygen carrier |        |       |         |          |           |
| A1       | 30     | 303.15| 1.01    | 792.74   | 0.00      | M1       | 1200   | 1473.15| 13      | 514.20   | 1238.99   |
| A2       | 152    | 425.15| 3.43    | 792.74   | 123.71    | M2       | 1226   | 1499.15| 11.6    | 461.33   | 1017.38   |
| A3       | 35     | 308.15| 13.5    | 792.74   | 5.02      | S1       | 30     | 303.15| 1.01    | 9.64     | 0.00      |
| A4       | 183    | 456.15| 13.4    | 739.50   | 155.54    | M2       | 31     | 304.15| 150     | 9.64     | 4.18      |
| A5       | 183    | 456.15| 13.5    | 739.50   | 155.54    | S2       | 185    | 458.15| 150     | 9.64     | 792.24    |
| A6       | 413    | 686.15| 13.5    | 739.50   | 397.37    | S3       | 182    | 598.15| 20      | 9.64     | 3157.84   |
| A7       | 1200   | 1473.15| 13      | 686.63   | 1309.80   | S4       | 180    | 733.15| 20      | 9.64     | 3081.50   |
| A8       | 497    | 770.15| 1.01    | 739.87   | 491.04    | S5       | 325    | 598.15| 20      | 9.64     | 3081.50   |
| A9       | 272    | 544.94| 1.01    | 739.87   | 249.32    | S6       | 470    | 743.15| 20      | 9.64     | 3040.01   |
| A10      | 266    | 539.15| 1.01    | 739.87   | 243.07    | S7       | 190    | 463.15| 1.7     | 9.64     | 2852.17   |
| A11      | 75     | 348.15| 1.01    | 739.87   | 45.71     | S8       | 31     | 304.15| 1.7     | 86.97    | 4.18      |
| Fuel     |        |       |         |          |           | Oxygen carrier |        |       |         |          |           |
| F1       | 30     | 303.15| 13      | 71.43    | 0.00      | M1       | 1200   | 1473.15| 13      | 514.20   | 1238.99   |
| F2       | 163    | 436.15| 13      | 71.43    | 219.96    | M2       | 1226   | 1499.15| 11.6    | 461.33   | 1017.38   |
| Exhaust  |        |       |         |          |           | S1       | 30     | 303.15| 1.01    | 9.64     | 0.00      |
| E1       | 1226   | 1499.15| 11.8    | 124.30   | 1691.73   | S1       | 190    | 463.15| 1.7     | 50.97    | 2852.17   |
| E2       | 761    | 1034.15| 1.01    | 124.30   | 976.25    | S1       | 190    | 463.15| 1.7     | 50.97    | 2852.17   |
| E3       | 680    | 953.15| 1.01    | 124.30   | 849.85    | S1       | 190    | 463.15| 1.7     | 50.97    | 2852.17   |
| E4       | 640    | 913.15| 1.01    | 124.30   | 788.41    | S1       | 190    | 463.15| 1.7     | 50.97    | 2852.17   |
| E5       | 517    | 790.15| 1.01    | 124.30   | 604.96    | S1       | 190    | 463.15| 1.7     | 50.97    | 2852.17   |
| E6       | 500    | 773.15| 1.01    | 124.30   | 580.09    | S1       | 190    | 463.15| 1.7     | 50.97    | 2852.17   |
| E7       | 128    | 401.15| 1.01    | 124.30   | 76.98     | S1       | 190    | 463.15| 1.7     | 50.97    | 2852.17   |
| E8       | 40     | 313.15| 1.01    | 124.30   | 23.56     | S1       | 190    | 463.15| 1.7     | 50.97    | 2852.17   |
| E9       | 40     | 313.15| 1.05    | 38.20    | 41.72     | S1       | 190    | 463.15| 1.7     | 50.97    | 2852.17   |
| E10      | 40     | 313.15| 1.05    | 86.09    | 8.54      | S1       | 190    | 463.15| 1.7     | 50.97    | 2852.17   |
| E11      | 35     | 308.15| 110     | 86.09    | 4.26      | S1       | 190    | 463.15| 1.7     | 50.97    | 2852.17   |

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Fig. 10  Schematic of syngas-fueled combined-cycle chemical-looping combustion power plant without the provision for carbon capture
These are then used partly to preheat the gaseous reactants and partly to generate a Rankine cycle to generate further electrical power. In the process of giving up the heat for the Rankine cycle, the oxygen-depleted air from the air reactor gets cooled down to ~120 °C and is fed to a chimney as it contains little CO₂. The exhaust from the fuel reactor to get cooled down to ~100 °C as it passes through the heat recovery systems. For the CCS mode of operation of the power plant, the exhaust stream (E6) is admitted into the flue gas conditioner, where it is finally cooled to 40 °C (E5). The condensed water vapor is separated (E6), and CO₂ gas (E7) is compressed to 110 bar (E8) at 35 °C if CCS is mandatory or let into the atmosphere through the air chimney when CCS need not be implemented as shown in Fig. 11.

Specific features of the layout that make it advantageous and future-ready are:

- The use of a combined-cycle operation to ensure high power output
- The use of a chemical-looping combustion mode of the burning of hydrocarbon fuel enables CO₂ capture as a built-in feature.
- The in-built feature of CO₂ capture enables the power plant to be ready for CCS when it becomes mandatory.
- By proper sizing of the major equipment, it is possible for the plant to be used with as widely different fuels as methane-rich natural gas (or shale gas) and CO/H₂-rich syngas (or biogas).

These aspects are elaborated further below:

A process flow diagram of the proposed power plant layout is shown in Fig. 10. Based on mass and energy balances discussed in the “Layout of natural gas-fired pressurized CLC” and “Layout of syngas fired pressurized CLC” sections, this has been developed for a 761 MWth plant when fired with natural gas and 800 MWth when fired with syngas. Major has now been identified, and the flow paths of the various material streams, such as air, hydrocarbon fuel, steam, and metal oxide, have been identified. Process flow sheeting calculations have been done for four different modes of operation, namely, with natural gas firing with and without CCS, and with syngas firing with and without CCS, and the pressure, temperature, and flow rate at several important locations in the flow paths have been calculated. These are summarized in Table 3. The fact that the same layout descriptors can be used for all four configurations highlights the multi-mode capability of the plant design. The two specific features, namely, dual fuel flexibility and readiness for CCS, engender small changes in the process stream flow paths. These have been identified in Fig. 10. Fuel switching from syngas to natural gas requires the fuel reactor exhaust stream to go directly from E3 to E5, thus bypassing the high pressure and the medium pressure turbines of the Rankine cycle system. There should be a corresponding bypassing of the relevant heat recovery steam generators by the water on the steam side. Switching from a without CCS mode (which is the current practice) to with CCS mode (which may be required in the future) requires the fuel reactor exhaust to be sent to a CO₂ compressor via an exhaust gas cooler to remove steam instead of being sent to the air chimney.

Finally, the overall efficiency of the power plant under various modes of operation is summarized in Table 3. Here, a listing is made of the various processes by which power is produced and those by which power is consumed for the four configurations.

Overall energy analysis is made for power plant layouts and net efficiency for each case is evaluated using Eq. (7) considering the energy penalties for gas compression and water circulation.

\[
\eta_{\text{Net}} = \frac{P_{\text{Net CC CLC}}}{m_{\text{fuel}} Q_{\text{LHV}}} \quad (7)
\]

In Eq. (7), \( \eta_{\text{Net}} \) is net efficiency of power plant, \( m_{\text{fuel}} \) is mass flow rate of fuel in kg/s, \( Q_{\text{LHV}} \) is lower heating value (kJ/kg) of the fuel and \( P_{\text{Net CC CLC}} \) is net power available for combined-cycle chemical-looping combustion power plant and \( P_{\text{Net CC CLC}} \) evaluated using Eq. (8)

\[
P_{\text{Net CC CLC}} = P_{\text{CO₂+Air+steam}} - P_{\text{Air comp}} - P_{\text{CO₂ comp}} - P_{\text{water pump}} \quad (8)
\]

In Eq. (8), \( P_{\text{CO₂+Air+steam}} \) is power produced (kW) by CO₂/ exhaust, air, and steam turbines, \( P_{\text{Air comp}} \) and \( P_{\text{CO₂ comp}} \) is, respectively, power consumed (kW) by air and CO₂ compressors and \( P_{\text{water pump}} \) is the power consumed (kW) to pump water.

It can be seen that with CCS, a net thermal efficiency of 52.13% is obtained with natural gas and 48.78% with syngas using Eqs. (7) and (8). In without CCS mode (without CO₂ compression), higher thermal efficiencies of 54.06 and 52.63% efficiencies are obtained. The lower difference between the natural gas and the syngas in without CCS mode is reflective of the higher rate of CO₂ generation in the latter per MWth of the fuel. The high efficiencies obtained in all these cases are partly due to combined-cycle operation, partly due to the use of supercritical boilers in the steam cycles, and partly due to the use of chemical-looping combustion as opposed to oxyfuel combustion. It may be noted that in these calculations, allowance has been made for non-idealities in the expansion, compression, and pumping processes involving fluid flow. Allowance has also been made for the power required to compress CO₂ to a pressure of
Fig. 11  Schematic diagram of multi-fuel compatible combined-cycle CLC-based power plant layout with provisions for, with CCS and, without CCS modes.
110 bar so that the nearly pure (purity > 99%) can be sent to a CO₂ sequestration/storage/reuse site. Thus, it can be concluded the above power plant, which offers operational flexibility coupled with high efficiency, has the necessary attributes of a future-ready power plant producing power from conventional hydrocarbon fuels in an environmentally friendly way.

**Assessment of possibility of dual fuel operation**

In order to assess the possibility of dual fuel operation of the pressurized CLC system, a detailed comparison of several parameters associated with the two systems is made in Table 4. It can be seen that most of the parameters are fairly similar; the operating temperatures of the fuel reactors are slightly different. Due to the exothermicity of the reduction reaction, the fuel reactor temperature is actually higher than that of the air reactor by about 26 °C with syngas, whereas it is lower by about 50 °C in the case of natural gas firing. Another difference between the two fuels is the calorific value and the fuel gas flow rate. Since syngas contains a significant amount of CO₂ as inert, its flow rate is higher and the flow rate of the exhaust gas from the fuel reactor is also higher. This, coupled with the higher exit temperature of the fuel reactor and lesser gas preheating requirement, results in a significantly higher amount of thermal power (105.64 MW for syngas vs 51.71 MW for natural gas) retained in the fuel reactor exhaust gas (see Figs. 6 and 9) which is available for powering a Rankine cycle. As can be seen from Table 4, the Rankine cycle for syngas has three turbines (a high-pressure turbine (150 bar), a medium pressure turbine (20 bar), and a low-pressure turbine (1.7 bar)) while the one for natural gas has a single low-pressure turbine with a turbine inlet pressure of 1.7 bar.

It can be seen that the major part of the power is produced by the low-pressure turbine in the syngas case and that its rating is roughly the same (33,216 to 29,396 kWₑ) as that of the low-pressure turbine in the natural gas case. The air reactor side parameters are nearly similar, and there is hardly a 5% change in the heat and mass flow rates of various streams. From the above reasoning, it can be argued that except for minor changes in the layout for the Rankine cycle parameters, the layouts of the syngas-fired and the natural gas-fired pressurized CLC power plants are similar, and a unified layout is produced in Fig. 11. In this common layout, natural gas firing requires the by-passing of the fuel reactor exhaust from E3 to E6 directly. Correspondingly, the steam side bypasses the high-pressure and the medium-pressure turbines and uses only the low-pressure turbine. The stream values corresponding to the unified layout are given in Table 1 for natural gas and Table 2 for syngas firing. These represent only minor departures from the normal settings and can be re-engineered without significant difficulty. This means that the thermodynamic compatibility between syngas and natural gas layouts for pressurized CLC can be achieved without significant retrofitting. The reaction engineering and transport phenomena considerations are similar to those already as our earlier work (Basavaraja and Jayanti 2015b). While the kinetics of pressurized oxidation/reduction reactors have not been fully worked out, it appears that the kinetics of all the reactions are slowed down in a similar way in the pressure ranges considered (Adanez et al. 2012; Garcia-Labiano et al. 2006). As demonstrated in our earlier work (Basavaraja and Jayanti 2015b), the fluidization and heat transfer compatibility conditions can be met by careful design of the relevant equipment. Thus, it appears that dual-fuel flexibility can be incorporated into pressurized CLC layout at the design stage itself. Table 4, work reported by Naqvi et al. (2007) for 697,545 kW thermal in put natural gas-fired pressurized CLC plant operates at 1200 °C (13 bar) at air reactor and 980 °C (11.6 bar) at fuel reactor. The oxygen carriers, operating pressure (at air and CO₂ turbines), compressor efficiency chosen is same as reported by Naqvi et al. (2007) for the natural gas-fired CC CLC plant layout calculations of the present work with 761,000 kW thermal input and the net efficiency is found to be 52.13% and which is comparable net efficiency reported.

### Table 3 Energy analysis of combined-cycle CLC-based power plants operating in with CCS and without CCS modes

| Power produced/consumed | Natural gas CC CLC | Syngas CC CLC |
|-------------------------|-------------------|---------------|
| **Power produced**      |                   |               |
| Air turbine (kWₑ)       | 596,064           | 544,302       |
| Carbon dioxide rich gas turbine (kWₑ) | 61,674 | 88,934 |
| Steam turbine power (kWₑ) | 29,396 | 39,253 |
| **Power consumed**      |                   |               |
| Air compression (kWₑ)   | 275,716           | 251,197       |
| CO₂ compression to 110 bar (kWₑ) | 14,721 | 30,842 |
| Water pump cost (kWₑ)   | 10                | 202           |
| Available power (kWₑ)   | 396,6877          | 390,248       |
| Thermal input (kWₑ)     | 761,000           | 800,061       |
| **Net efficiency (%)**  | 52.13             | 52.63         |
by Naqvi et al. (2007). The detailed analyses of CC CLC reactor operating conditions, power produced at gas and steam turbines, power consumed for air and CO₂ compression, net efficiencies are shown in Table 4.

**Future-readiness of the layout**

The lifetime of a thermal power plant is in excess of 50 years. While carbon capture and sequestration are not mandatory right now, it is possible that it may become so in a decade or two. In this scenario, it is necessary to build power plants that are future-ready, i.e., which can be made to operate in a case where CCS becomes mandatory. Extensive retrofitting of the power plant to accommodate carbon capture may not be possible and should be avoided. From the future-compatibility point of view, a chemical-looping combustion-based power plant is clearly the superior design. CLC plants can work in either with CCS mode or without CCS mode with very few

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**Table 4** Overall energy analysis for combined-cycle CLC

| Variants                                    | Syngas layout | Natural gas layout | Natural gas layout (Naqvi et al. 2007) |
|---------------------------------------------|---------------|--------------------|--------------------------------------|
| Air reactor operating temperature (°C)      | 1200          | 1200               | 1200                                 |
| Fuel reactor operating temperature (°C)      | 1226          | 1150               | 980                                  |
| Temperature of 13 bar pressure oxygen       | 1132          | 1132               | 1140                                 |
| Temperature of 1 bar pressure oxygen        | 497           | 497                | 492                                  |
| CLC fuel reactor exhaust gas turbine inlet temperature (°C) at pressure 11.6 bar | 1226          | 1150               | 980                                  |
| CLC fuel reactor exhaust exit temperature (°C) at 1 bar (at 92% isentropic efficiency) | 761           | 666                | 533                                  |
| Mass flow rate of depleted air at air turbine inlet/exit after addition of coolant air in CC CLC (kg/s) | 739.87        | 810.43             | 768                                  |
| Mass flow rate of fuel reactor exhaust gas at exhaust gas inlet/exit in CC CLC (kg/s) | 124.30        | 75.1               | 70.5                                 |
| CLC air turbine inlet/exit gas composition (wt%) | 82.7% N₂ and 17.3% O₂ | 82.85% N₂ and 17.15% O₂ | 83% N₂ and 17% O₂                     |
| CLC exhaust gas turbine inlet/exit gas composition (wt%) | 30.63% H₂O and 69.37% CO₂ | 45.32% H₂O and 54.68% CO₂ | 45.32% H₂O and 54.68% CO₂              |
| CO₂ purity (wt%) for CCS | > 99          | > 99               | 99                                   |
| **Power produced**                          |               |                    |                                      |
| Air turbine (kWₑ)                           | 544,302       | 596,064            | 477,000                              |
| Carbon dioxide rich gas turbine (kWₑ)       | 88,934        | 61,674             | 53,600                               |
| Steam cycle                                 |               |                    |                                      |
| Steam turbine power (kWₑ)                  | 39,253        | 29,396             | 101,000                              |
| **Power consumed**                          |               |                    |                                      |
| Air compression (kWₑ)                       | 251,197       | 275,716            | 243,000                              |
| CO₂ compression (kWₑ) (compressor efficiency 85%) | 30,842 (to 110 bar) | 14,721 (to 110 bar) | 15,400 (to 200 bar)                   |
| Water circulation cost (kWₑ) (pump efficiency 75%) | 202 (1 to 150 bar) | 10 (1 to 1.7 bar) | -                                    |
| **Available power (kWₑ)**                   |               |                    |                                      |
| Thermal input (kW)                          | 390,248       | 395,267            | 364,000                              |
| Net efficiency (%) (with CO₂ compression)   | 48.78         | 52.13              | 52.18                                 |
changes required to make the switch. No special efforts or equipment will be necessary to enable CO₂ capture. If CCS is not needed, then the flue gas from the flue gas conditioner need not be compressed and can be sent directly to the chimney. When CCS is required, then this flue gas can be diverted to a CO₂ compressor. In the case of a power plant based on oxyfuel combustion, more extensive changes are required as an air separation unit is required for generating pure oxygen. While studies (Jayanti et al. 2012) indicate that retrofitting of atmospheric air combustion coal-fired boilers for working in oxyfuel mode is possible, the case for pressurized oxyfuel combustion may be different because flue gas compression and recirculation are necessary. In the case of pressurized oxyfuel combustion, the flow rates and composition of the exhaust gas are significantly different from those under air combustion; this may warrant changes in the turbine and heat recovery systems.

**Conclusion**

In the present study, an analysis of two modes of operation of two gas-fired pressurized chemical-looping combustion-based power plants has been made. The principle conclusions drawn from the study is given:

- From the detailed mass and heat balances and thermodynamic analysis, power plant layouts have been prepared for pressurized CLC-based power plants for fuels, natural gas, and syngas. The oxidation reaction in the fuel reactor of a CLC plant can be exothermic (for example, for syngas) or endothermic (for example, for natural gas) depending on the fuel used. This aspect has been thoroughly examined in the present study from the point of view of the power plant layout.

- Two modes of power plant operation, one not requiring CCS (the present-day mode) and one requiring CCS (in a future scenario), have been considered within the ambit of a pressurized CLC-based power plant. The net efficiency of the plant, after making allowance for thermodynamic irreversibility and CO₂ compression, has been found 52.13% with natural gas and 48.78% with syngas. The net efficacy gain of 2% with natural gas-fired pressurized CLC plant and 4% with syngas fired pressurized CLC plant found without CO₂ compression. The potential for fuel flexibility using CLC-based power plants has been explored by developing layouts for natural gas and syngas. It is shown that since CLC generates CO₂-rich flue gas (contains CO₂ and water vapor only), which is ready for sequestration (carbon capture mode), the power plant can be readily operated in a dual mode with a loss of 2% of net thermal efficiency with natural gas and up to 4% for syngas.

- A single layout of future-ready power plants offering in-built CO₂ capture, high efficiency, and fuel flexibility has been developed for power generation using gaseous hydrocarbon fuels.

- Oxyfuel combustion-based power plants are technically mature in terms of proven commercial-scale operation of new elements. They also have competitive net thermal efficiency if pressurized steam moderated oxyfuel combustion (SMOC)-based oxyfuel combustion is used. However, they make a poor choice in terms of future readiness of fuel/equipment flexibility and may warrant significant retrofitting effort to make them CCS-compatible. CLC plants can work in either with CCS mode or without CCS mode with very few changes required to make the switch.

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