

CRANFIELD UNIVERSITY

Xiaoyu Wei

EVALUATION OF IMPROVEMENTS ON TECHNO-ECONOMIC
PERFORMANCE OF A SUPERCRITICAL OXY-FUEL
COMBUSTION POWER PLANT

School of Water, Energy and Environment
PhD in Energy and Power

Doctor of Philosophy
Academic Year: 2017 - 2020

Supervisor: Dr Dawid Hanak
Associate Supervisor: Prof Vasilije Manovic
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the degree of Doctor of Philosophy

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ABSTRACT

To meet the global target for lowering greenhouse gas emissions and preventing climate change, the power sector has to be decarbonised by 2050. Since coal is projected to play a critical role in the future energy portfolio, carbon capture and storage (CCS) technology has taken on the role of decarbonisation. To find the ways potentially improve defects of the state-of-art oxy-fuel combustion power plant regarding worse techno-economic performance, the oxy-fuel combustion power plant coupling with recuperated supercritical carbon dioxide (sCO₂) cycle has the potential to surpass the state-of-art oxy-fuel combustion power plant. Its net electricity efficiency and levelised cost of electricity (LCOE) is 29.73% and 97.7 €/MW_{el}h, respectively, at operating conditions of 593°C and 240 bar. Further study of chemical looping combustion (CLC) power plant with recompression sCO₂ power cycle achieved 35.49% of net electricity efficiency and 96.8 €/MW_{el}h of LCOE for the manganese ore as the natural oxygen carrier. Conversely, an LCOE of 109.2 €/MW_{el}h was obtained owing to selecting the manufactured oxygen carrier of nickel oxide. Particularly worth mentioning is the excellent decarbonisation ability by fuelling the biomass in the two processes with -1255 gCO₂/kW_{el}h and -1066 gCO₂/kW_{el}h (Mn₃O₄ case) of specific carbon dioxide (CO₂) emission, respectively. However, it showed significantly higher LCOE with around 2.3%_{HHV} and 3.8%_{HHV} (Mn₃O₄ case) of net efficiency penalty, respectively, compared with the coal-fuelled cases. In the emission trading system with the uncertainty of carbon tax, the two processes fuelled by biomass have the potential to achieve lower costs than the coal-fuelled cases. Finally, the probability assessment is conducted in the proposed cases, showing a higher cumulative probability of LCOE in the coal-fuelled CLC case than in the oxy-coal case. Hence, this study revealed that technologies of CLC and the sCO₂ power cycle have the potential to improve the techno-economic performance of the state-of-art oxy-fuel combustion technology.

Keywords: Oxy-fuel combustion, chemical looping combustion, supercritical CO₂ power cycle, techno-economic assessment, probabilistic analysis

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LIST OF NOMENCLATURE

A_{HE}	Heat exchange surface area	m^2
C_C	BEC of the supercritical CO ₂ compressor	M€
C_{CT}	BEC of the cooling tower	M€
C_{cycle,CO_2}	BEC of the sCO ₂ power cycle	M€
C_E	BEC of the sCO ₂ expander	M€
C_{EG}	BEC of the electric generator	M€
C_{fix}	Fixed Carbon	mole
C_{HE}	BEC of the heat exchanger	M€
$e_{CO_2,capture}$	Specific CO ₂ emission of the power plant with CCS	gCO ₂ /kW _{el} h
$e_{CO_2,ref}$	Specific CO ₂ emission of the power plant without CCS	gCO ₂ /kW _{el} h
e_{CO_2}	Specific CO ₂ emission	gCO ₂ /kW _{el} h
$i_{P\&C}$	Piping and integration cost indicator	%
$LCOE_{CCS}$	Levelised cost of electricity with CCS	€/MW _{el} h
$LCOE_{CC}$	Levelised cost of electricity with carbon capture	€/MW _{el} h
$LCOE_{ref}$	Levelised cost of electricity without CCS	€/MW _{el} h
$\dot{m}_{AE,Com/Exp}$	Equivalent mass flow rate of air at the inlet to the compressor/expander	kg/s
$\dot{m}_{CO_2}$	CO ₂ emission rate	g/a
$\dot{m}_{CO_2,Com/Exp}$	Equivalent mass flow rate of CO ₂ at the inlet to the compressor/expander	kg/s
$\dot{m}_{fuel}$	Mass flow rate of the fuel	kg/s
m_{ox}	Fully oxidized form of the oxygen carrier	kg
m_{red}	Fully reduced form of the oxygen carrier	kg
$p_{AE,out}$	Equivalent air outlet pressure	bar
p_{HE}	Highest pressure of the heat exchanger	bar
p_{in}	Expander/compressor inlet pressure	bar
P_{ASU}	ASU power consumption	MW _{el}
P_{CPU}	CPU power consumption	MW _{el}
P_{FAN1}	Flue gas circulation fan power consumption	MW _{el}
P_{FAN2}	Main fan power consumption	MW _{el}
P_G	Gross power output	MW _{el}

$\sum P_{AUX}$	Total power consumption of the auxiliary equipment	MW _{el}
$\dot{Q}_{CT}$	Heat duty of the cooling tower system	MW _{el}
r	Discount rate	%
R_O	Oxygen transport capacity	
T_{in}	Expander inlet temperature	°C
T_{FR}	Fuel reactor temperature	°C
T_{AR}	Air reactor temperature	°C
$\dot{W}_{net}$	Net power output	kW _{el}
$X_{char,FR}$	Char conversion efficiency in fuel reactor	%
$\eta_{i,Com}$	Compressor isentropic efficiency	%
$\eta_{i,Exp}$	Expander isentropic efficiency	%
η_{net}	Net electricity efficiency of the entire power plant	%
$\rho_{in,Air}$	Air density at the inlet of compressor/expander	kg/m ³
ρ_{in,CO_2}	CO ₂ density at the inlet of compressor/expander	kg/m ³
ρ_{out,CO_2}	CO ₂ density at the outlet of compressor/expander	kg/m ³
$\rho_{out,Air}$	Air density at the outlet of compressor/expander	kg/m ³

LIST OF ABBREVIATIONS

A	Area
AC	Cost of CO ₂ avoided
AR	Air reactor
ASPEN	Advanced System for Process Engineering
ASU	Air separation unit
BEC	Bare erected cost
BECCS	Bioenergy with carbon capture and storage
BFB	Bubbling fluidized bed
BOP	Balance of plant
C	Compressor
CC	Cost of CO ₂ captured
CCS	Carbon capture and storage/sequestration
CDCLC	Coal direct chemical looping combustion
CF	Capacity factor
CFB	Circulating fluidized bed
CFPP	Coal-fired power plant
CL	Chemical looping
CLC	Chemical looping combustion
CLOU	Chemical looping with oxygen uncoupling
COE	Cost of electricity
COP	Compressor outlet pressure
COP21	The 21 st session of the Conference of the Parties
CPU	Compression and purification unit / CO ₂ purification unit
DCOALIGT	Institute of Gas Technology coal density model
DECHEMA	German Society for Chemical Apparatus
EO	Equation oriented
EOM	Enterprise Operations Management
EOR	Enhanced oil recovery
EPRI	Electric Power Research Institute
ETS	Emission Trading System
EU	European Union

EU ETS	European Union Emission Trading System
FCF	Fixed charge factor
FGD	Flue-gas desulfurization
FOM	Fixed operating and maintenance cost
FR	Fuel reactor
GRACE	Grangemouth Advanced CO ₂ Capture
GCCSI	Global Carbon Capture and Storage Institute
GHG	Greenhouse gas emissions
HCOALGEN	General coal enthalpy model
HHV	Higher heating values
HNIRI	Hokkaido institute of industry
HRSG	Heat recovery steam generator
HTR	High-temperature recuperator
HVAC	Heating, ventilation and air conditioning
HX	Heat exchanger
ICB-CSIC	Instituto de Carboquímica in the Consejo Superior de Investigaciones Científicas Institute of Carbochemistry in Spanish National Research Council
ICLAS	Integrated chemical looping air separation unit
IEA	International Energy Agency
IFRF	International Flame Research Foundation
IGCC	Integrated Gasification Combined Cycle
iG-CLC	In situ gasification CLC
IL-ICCS	Illinois Industrial CCS
IPCC	Intragovernmental Panel on Climate Change
IRR	Internal rate of return
LCOE	Levelised cost of electricity
LK-PLOCK	Lee-Kesler-Plöcker equation of state
LMDT	Log mean differential temperature across the heat exchanger
LTR	Low-temperature recuperator
MC	Main compressor
MDEA	Methyldiethanolamine
MEA	Monoethanolamine
MIT	Massachusetts Institute of Technology

NASA	National Aeronautics and Space Administration
NETL	National Energy Technology Laboratory
NPV	Net present value
NRE	Non-recoverable engineering
OC(s)	Oxygen carrier(s)
OEM	Original equipment manufacturers
O&M	Operations and maintenance
PC	Primary cooler
PFBC	Pressurized fluidized bed combustion
PHX	Primary heat
PR-BM	Peng Robinson cubic equation of state with the Boston-Mathias alpha function
Q	Transferred heat in the heat exchanger
RC	Recycle compressor
SC	Steam cycle
sCO ₂	Supercritical carbon dioxide
SCR	Selective non-catalytic reduction
SFC	Specific fuel cost
SM	Sequential module
SNCR	Selective catalytic reduction
T	Turbine
TCR	Total capital requirement
TEA	Techno-economic assessment
TIT	Turbine inlet temperature
TRL	Technology readiness level
TS&M	Transport, storage, and monitoring
U	Universal heat transfer coefficient
USDOE	The United States Department Of Energy
VOM	Variable operating and maintenance cost
WGS	Water-gas shift
WP	Wood pellet

1 Introduction

1.1 Background

The report from the Intergovernmental Panel on Climate Change confirms that anthropogenic activities have contributed to climate change with an over 95% probability [1]. Carbon dioxide (CO_2) emissions as a central component of the greenhouse gases are recognised as the major reason influencing climate change, with 41% CO_2 emission from electricity and heat generation, which has over 65% of electricity and heat generation emissions from coal [2]. The average concentration of CO_2 in the atmosphere has increased from 280 ppm to 414 ppm from the Industrial Revolution to February 2020 (Figure 1-1). The annual rate of increase for atmospheric CO_2 over the past 60 years is around 100 times that at the end of the last ice age (previous natural increases) [3], which establishes the greenhouse effect. Even if CO_2 emissions are stopped, the effects of global warming will continue for centuries [1].

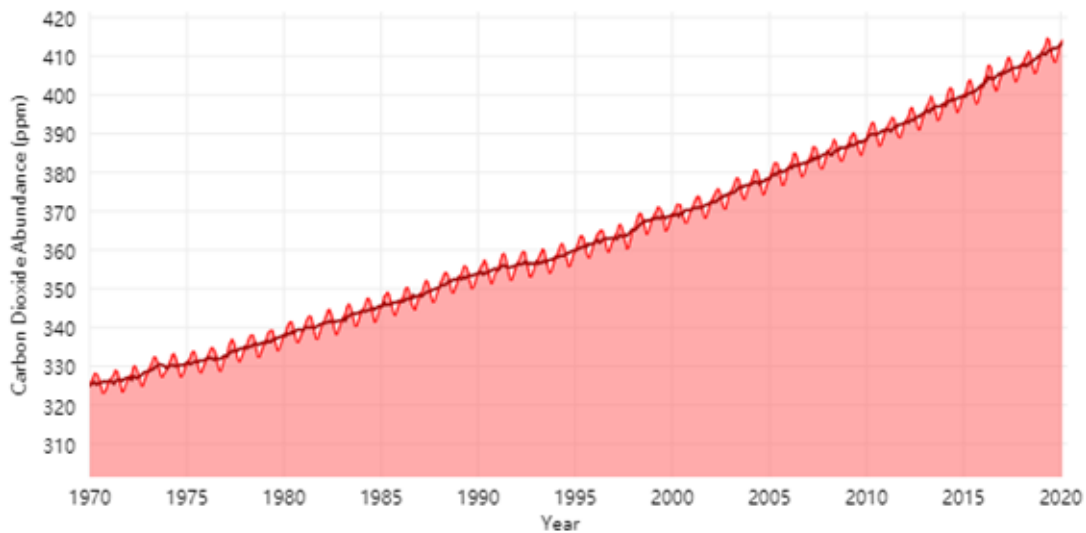


Figure 1-1 Trend of global average CO_2 concentration [3]

In the 1860s, physicist John Tyndall recognised the Earth's natural greenhouse effect, and a slight change in the composition of the atmosphere can cause climate change. In 1896, Svante Arrhenius, a Swedish scientist, was the first to predict that

the surface temperature could be substantially altered through the greenhouse effect that is blamed for changes in the levels of CO₂ in the atmosphere. Meanwhile, the heat-trapping characteristics of CO₂ and other greenhouse gases were demonstrated with their ability to affect infrared energy transmission through the atmosphere. There is no doubt that the increase of greenhouse gases inevitably leads to the warming of the earth in response. Modern evidence and ancient evidence show that the global climate is influenced by variations of greenhouse gas concentration [4].

Climate change is defined as *“a change of climate which is attributed directly or indirectly to human activity that alters the composition of the global atmosphere and which is in addition to natural climate variability observed over comparable time periods”*, which distinguishes ‘climate change’ attributable to human activities altering the atmospheric composition and ‘climate variability’ attributable to natural causes [5]. The United Nations Framework Convention on Climate Change describes that changes in solar radiation, changes in the earth's orbit, volcanic activity, changes in the atmosphere and ocean circulation are natural factors attributed to global climate change. Moreover, anthropogenic activities since the Industrial Revolution are currently the major causes of global warming as the primary phenomenon of climate change, including anthropogenic greenhouse gases emissions, other emissions from human life, land use, and urbanization.

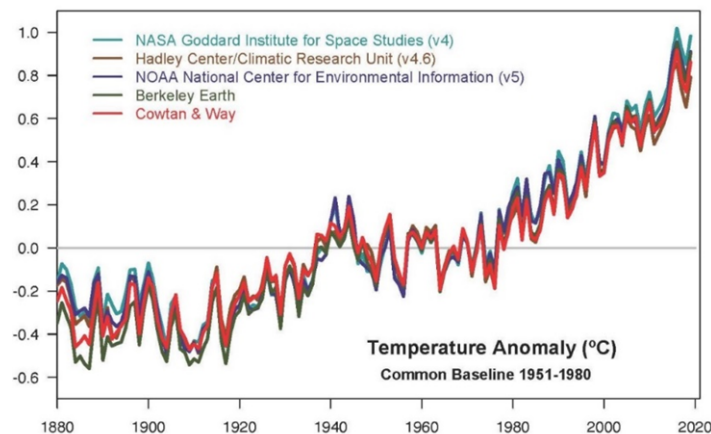


Figure 1-2 Yearly temperature anomalies from 1880 to 2019 [6]

The anthropogenic greenhouse gas emissions (GHG) have resulted not only in that the global average surface temperature during 2006 to 2015 is 0.87°C higher than the average value during 1850 to 1900 [7] (Figure 1-2), but also in global temperature rise, sea level rise, warming oceans, shrinking ice sheets, glacial retreat, decreased snow cover, declining arctic sea ice, extreme events and ocean acidification [8] with following pieces of evidence. It shows that most of the warm-up earth surface has occurred in the past 35 years, and after 2014, the six warmest years is on record, such as the warmest year of 2016 [9,10]. If it continues on this upward trend, the global average temperature will potentially rise by 1.5°C after 20 years compared with the level of the pre-industrial period [7]. Since 1969, ocean temperature has increased more than 0.33°C observed by the National Oceanic and Atmospheric Administration [11]. Climate experiment from the National Aeronautics and Space Administration (NASA) shows that, on average, 286 billion tons of ice were lost per year in Greenland between 1993 and 2016. Similarly, 127 billion tons of ice per year in Antarctica were lost with tripled loss rate in the last decade [12]. Glaciers are retreating in most places around the world, such as the Alps and the Himalayas [13]. In the past 50 years, the amount of snow cover in the Northern Hemisphere has melted earlier [14]. In the last few decades, the extent and thickness of Arctic sea ice have been rapidly reduced [15]. Figure 1-3 shows the average extent of Arctic sea ice each September since the end of the 1970s [16]. Melting ice sheets and glaciers is a primary factor of sea level rise. In the last century, the global sea level has risen around 8 inches with a rate approximately twice over the last two decades than the last century [17]. Figure 1-4 shows that the sea level increases with a change rate of 3.3 millimetres per year since 1993, as observed by satellites [18]. In the United States, extreme high-temperature events have increased, low-temperature events have decreased, and heavy rainfall events have increased [19]. Since the Industrial Revolution, the surface acidity of seawater has increased by more than 30% [20,21], with about a 2 Gt each year increase in the CO_2 content of the oceans [22]. The effect of global warming is considerable. At least 35,000 people died in the European heatwave in 2003 [23].

Global tropical cyclones will be more intense, average rainfall rates of future tropical cyclones will be higher, and storm surge levels will be higher, such as Harvey and Irma in 2017 [24,25]. Several indirect harms affect animals and plants that are endeavouring to adapt to new environments, such as many creatures are moving north or into deeper waters to stay alive with habitat change [26]. As the above events have shown, the public must reduce, even cut, CO₂ emissions. The impacts of climate change on humans and nature may have positive or negative consequences for livelihoods, social and cultural assets, ecosystems and species, infrastructure and economic, services, health and well-being [1].

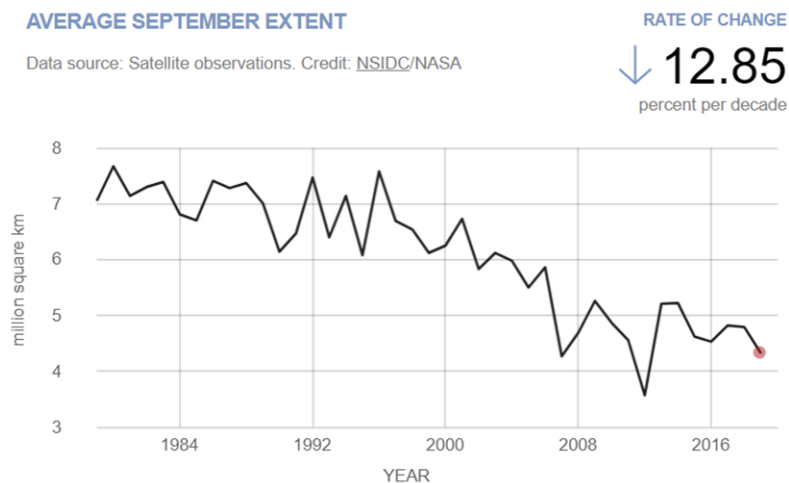


Figure 1-3 Average September extent of Arctic sea ice since 1979 [27]

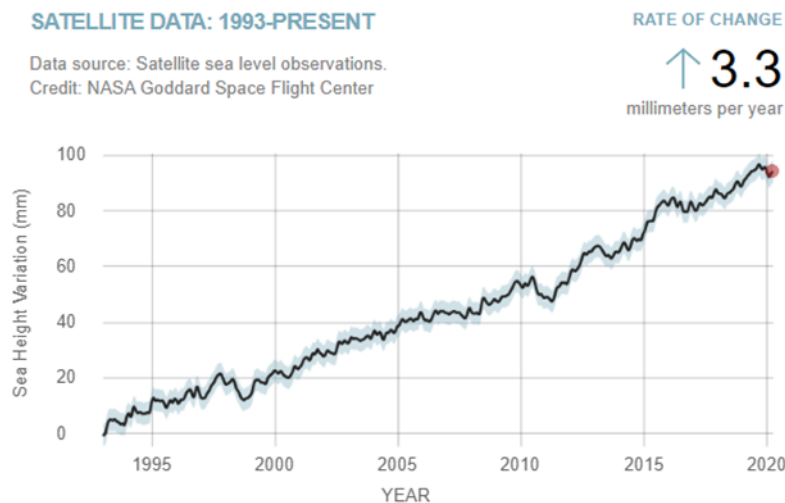


Figure 1-4 Sea level from 1993 to 2020 [28]

Coal was the fuel of economic growth in the industrial era. Today, cheap and abundant coal is the world's leading power source. As climate change worsens, many countries worldwide are phasing out coal-fired power plants and setting a timetable for reducing emissions. However, giving up coal will not be easy. In 2015, 195 countries adopted the Paris Climate Agreement, pledging to limit the global average temperature rise to 2°C. To achieve this, global carbon dioxide emissions would have to be cut by 45 per cent. Coal burning is the single largest source of carbon dioxide emissions from human activity, and many countries have begun to set timetables for reducing emissions. For example, on January 26, the Coal Committee appointed by the German government recommended that the government close all coal-fired power plants in the country by 2038 at the latest in order to achieve the emission reduction target. The British government announced in 2015 that it planned to phase out all coal-fired power plants by 2025. A detailed plan was introduced in January 2018 to limit CO₂ emissions per kilowatt of electricity to a maximum of 450g. French President Emmanuel Macron also announced at the World Economic Forum in January 2018 that he would close all coal-fired power plants in the country by 2021.

1.2 Motivation

According to International Energy Agency (IEA) [2,29], higher CO₂ emissions in electricity and heat generation (around 42% of total CO₂ emissions) mainly resulted from coal combustion (around 71% of CO₂ emissions from electricity and heat generation). There is 10159646.0 GWh of electricity generated by coal, accounting for around 39% of total power generation. Therefore, these sectors are seen as one with the most significant potential for decarbonisation. There has been a shift in recent years in terms of where to apply CCS technologies, and the focus is not only on coal-fired power plants (CFPP), but also on industrial applications (iron and steel, cement, glass and other industrial heat users, and chemical and refineries) now. These vital industries and power generation are crucial to developing global infrastructure and currently account for 64.9% of global CO₂

emissions, so decarbonising them together is just as important. To achieve emission reduction targets, low carbon technologies should be implemented for achieving carbon neutrality by 2050, which can meet the medium-term goal set in the Paris Agreement. Carbon capture and storage (CCS) is an essential technology for decarbonisation, which is predicted for reducing almost one-fifth of global CO₂ emissions across the industry sector in the period to 2060 [30]. To achieve their long-term goals, CCS needs to develop faster. In the IEA Clean Technology Scenario, CFPP without CCS will become unsustainable by the middle of the century, raising the risk that plants built in the short term will become stranded assets. Any new coal plant should be equipped with CCS technology. Promoting research into techno-economic performance for CCS equipped CFPPs under tougher climate targets could expand their applications. In addition, using CCS in power generation is more complicated than in industry because the capacity of the CO₂ capture facilities required for power generation is much greater, which has uncertainty on the scale expansion and takes a longer time to expand to the required scale. However, the main challenges of integrating CCS technologies with power plants are high capital costs, high operating costs and large energy penalties. For instance, at a conventional pulverized coal power plant, the post-combustion CO₂ capture with a monoethanolamine (MEA) scrubber leads to a 40–95% increase in cost of electricity due to high capital costs, high operating costs [31] and a near 10% points reduction for system net efficiency [32]. Pre-combustion CO₂ capture was characterised with a 5-9% points reduction on system net efficiency than MEA process, but it has to face commercialization challenge due to gasifier and air separation unit (ASU) achieving around 1950 €/kW of capital intensive [31]. Therefore, it is necessary to make breakthroughs in the research field of CCS and apply it into the power generation sector, such as applying cost-effective CO₂ capture materials, equipping advanced power cycle systems and optimizing the process.

Oxy-fuel combustion technologies can capture nearly 100% of CO₂ from fuel combustion. This is achieved via firing solid fuels with high purity oxygen (O₂), which, to some extent, advantages over pre-combustion and post-combustion CO₂ capture technologies. The oxygen source could be supplied from ASU, the oxygen transport membrane, or the CLC technology's oxygen carrier. From the Massachusetts Institute of Technology (MIT)'s study for oxy-combustion with the cryogenic air separation unit, its cost of electricity (COE) increase by nearly 46% [33]. Oxygen transport membrane technology could achieve a higher efficiency of oxygen production than an ASU. However, sizeable auxiliary power is consumed by an energy-intensive step from air compression units. Energy analysis usually uses system efficiency as an evaluation indicator with energy transfer, including work transfer, heat transfer and energy associated with mass transfer. Comparisons of energy and exergy analyses for a coal-fired oxy-fuel combustion system employed the pressurized fluidized bed (37% of the exergy loss) and a CLC system (only 11.6% of the exergy loss) have been conducted, indicating CLC is conspicuously more efficient [34]. Energy is composed of exergy and anergy. Exergy represents the useful work of a system. When the system is reversible, exergy is equal to the maximum useful work of the system. Exergy loss represents the degree of irreversibility of the system and the amount of useful work lost. Anergy represents the part of the energy that cannot be used during the energy transformation process. Accordingly, CLC could be a substitution for oxy-fuel combustion technologies. In addition, the net efficiency of the CLC technology (42.01%) [35] shows to be more efficient for CO₂ capture than that of pre-combustion capture technology based on physical absorption (between 36%-39%) [36] and oxy-fuel combustion capture technology (37.9%) [37]. Moreover, employing advanced thermodynamic cycles is a way to improve the techno-economic performance for oxy-fuel combustion and CLC power plants. It is worth noting that the supercritical CO₂ (sCO₂) power cycle is a novel technology by virtue to its compact equipment size and layout, and high thermal efficiency at moderate turbine inlet temperature (TIT) range (450-750°C) due to low compression work

[38]. The studies from Electric Power Research Institute illustrate that the net thermal cycle efficiency of the sCO₂ power cycle achieved 53.1% over that of the AUSC steam Rankine cycle with 48.8% [39].

1.3 Aim and objectives

This study aims to investigate potential improvements in the techno-economic feasibility of the state-of-art oxy-fuel combustion power plants. In the first proposed system, it is hypothesised that replacing the conventional steam Rankine cycle with the recuperated sCO₂ power cycle in advanced oxy-fuel combustion power stations will improve their techno-economic performance. In the second proposed system, CLC for reducing higher energy penalty of ASU and more efficient recompression sCO₂ power cycle is integrated and investigated. In addition, from the concept of Bioenergy with carbon capture and storage (BECCS), the biomass as the fuel is assessed in the above two proposed systems for their techno-economic feasibility and decarbonisation potential. The above-proposed systems are developed and evaluated in Aspen Plus™ for thermodynamic performance. Moreover, the economic feasibility is developed in Microsoft Excel. Then, the sensitivity analysis is conducted for demonstrating the effect of the critical design assumptions of the considered cases. Finally, the feasibility of the considered cases is benchmarked against the reference of conventional CFPP and the state-of-art oxy-fuel combustion power stations.

To achieve the aim, the following objectives were established:

1. Develop and validate the process models associated with oxy-fuel combustion, CLC and sCO₂ power cycles in Aspen Plus™.
2. Integrate the proposed models. The first integrated model replaces the conventional steam Rankine cycle by recuperated sCO₂ power cycle in the state-of-art oxy-fuel combustion power plant. The second integrated model is to integrate the CLC system with recompression sCO₂ power cycle.

3. Estimate the thermodynamic and economic performance of developed processes, and compare them with the reference oxy-fuel combustion case.
4. Adopt a probabilistic methodology for demonstrating the economic performance of the oxy-fuel combustion and CLC power plants integrated with the sCO₂ power cycle, respectively.

1.4 Structure of the thesis

The interconnections of the chapters are presented in Figure 1-5.

In Chapter 1, the aim and objectives of the research were provided, resulting from the background information and motivation for this PhD thesis.

Chapter 2 presents the literature review of CCS with the main focus on oxy-fuel combustion, CLC and the sCO₂ power cycle. It also reviews the progress in the BECCS, which is a comparable technology with CCS.

Chapter 3 introduces the methods employed in this thesis, including the software selection, the techno-economic assessment and probabilistic assessment. The primary software Aspen PlusTM and Microsoft Excel is introduced. The techno-economic assessment provides detailed thermodynamic and economic performance information, including the univariate sensitivity analysis, applied in Chapter 4 and Chapter 5. Notably, the equations for the capital cost of the sCO₂ power cycle is highlighted. Finally, the economic probabilistic assessment for the two proposed systems is conducted based on the same critical economic design assumptions applied in Chapter 6.

Chapter 4 analyses the techno-economic performance of the state-of-art oxy-fuel combustion power plant by replacing the conventional steam cycle with recuperated sCO₂ power cycle for potential improvement in efficiency and cost. In addition, biomass is used as the fuel to demonstrate its negative emission capability and cost-benefit under the carbon tax.

Chapter 5 demonstrates the techno-economic feasibility of the CLC power plant with the recompression CO₂ power cycle, aiming for reducing high energy penalty on ASU and high LCOE. Similarly, the biomass is fuelled for assessing the feasibility of the concept of BECCS in the targeted systems.

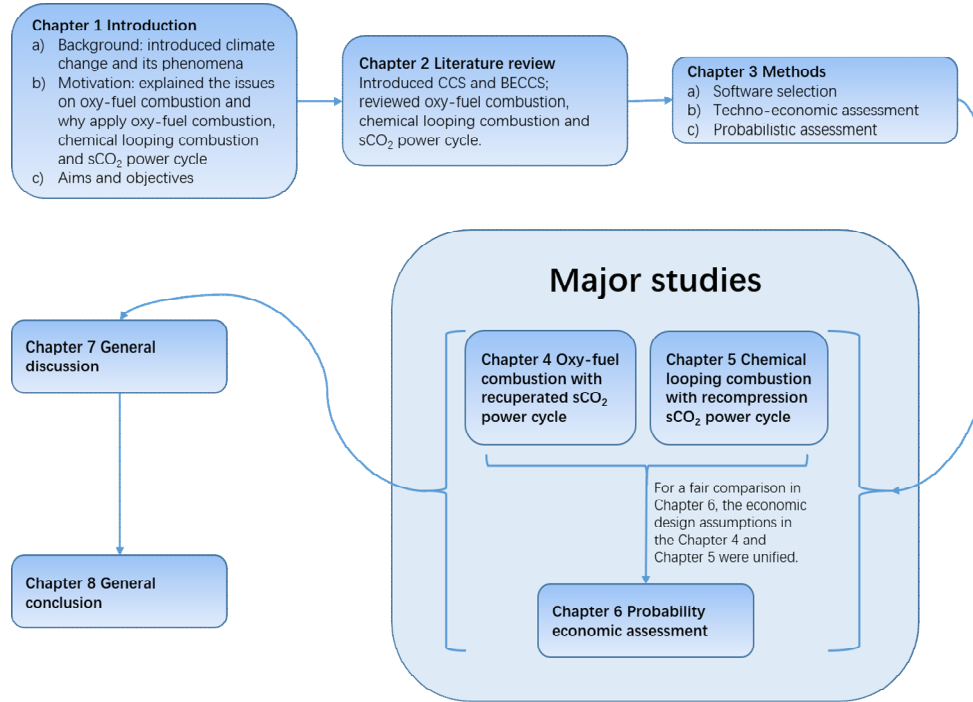


Figure 1-5 Interconnections of chapters of the thesis

In Chapter 6, the probability economic assessment of the systems proposed in Chapter 4 and Chapter 5 is carried out to analyse the economic performance under the influence of multi-parameter uncertainty.

In Chapter 7, the results of the research are compared and discussed with literature.

Chapter 8 gives a detailed and comprehensive summary results of this thesis. In addition, the suggestions were put forward for future development.

1.5 Dissemination from the PhD thesis

1.5.1 Publications

Wei X., Chen Y., Mennatallah L., Cann D. The different experience PACT facility can bring to you [Blog]. 27 September 2019. Available at:
https://ukccsrc.ac.uk/blog_posts/the-different-experience-pact-facility-can-bring-to-you/

Wei X., Manovic V., Hanak DP. [Techno-economic assessment of coal- or biomass-fired oxy-combustion power plants with supercritical carbon dioxide cycle](#). Energy Conversion and Management. October 2020; 221: 113143.

Wei X., Hanak DP. Effect of oxygen carrier selection on techno-economic performance of solid-fuelled chemical looping combustion with supercritical carbon dioxide power cycle [in preparation]. Cranfield University.

Wei X., Hanak DP. Probability economic assessment of a supercritical oxy-fuel combustion power plant [in preparation]. Cranfield University.

1.5.2 Presentation

Wei X. Evaluation of improvements on techno-economic performance of a supercritical oxy-fuel combustion power plant [presentation]. Research student seminar. Cranfield University, Cranfield, Bedford, UK. 2020.

2 Literature review

2.1 CO₂ capture technologies for coal-fired power plants

CCS technology is a process of capturing CO₂ from the flue gas and storing it in a way that prevents it from re-entering the atmosphere. It is expected to play a critical role in meeting the targets of preventing global warming set by the Intergovernmental Panel on Climate Change (IPCC) [40] and the twenty-first session of the Conference of the Parties (COP21) [41], expecting to achieve net-zero CO₂ emissions in the energy sector until 2070 [42]. It was predicted that coal-fired electricity generation shares less than 10% of the total electricity generation by 2050. Of these, 20% are equipped with carbon capture, utilisation and storage. As of April 23 2021, 44 countries and the European Union have pledged to achieving the net-zero emission target, occupying around 70% of global CO₂ emissions and gross domestic product [43]. The UK is now halfway to its 2050 net-zero emission target with a 51% GHG emission reduction from 1990 to 2020 [44]. In the US, there is a 50%-52% reduction in emissions by 2030 following its committed 2050 carbon neutral target. Uruguay's 2030 target is under discussion, which is the earliest carbon-neutral pledge. Sweden has legislated its 2045 target, which is the earliest commitment enshrined in law. In addition, the carbon-neutral goals of some countries or regions [45] are listed in Table 2-1.

Fossil fuels now supply most of the world's energy and are expected to continue for at least a few decades. CFPPs are the main source of CO₂ emissions, and therefore, there is an urgent need to reduce carbon emissions through CCS technologies. The most mature CCS technologies include pre-combustion, post-combustion and oxy-fuel combustion, according to the different locations where CO₂ is captured. The utilization and storage of CO₂ could be a solution to stabilize the GHG concentration in the atmosphere and will play a significant role in tackling climate change [46]. For the sake of continually use fossil fuels in an effective and environmentally sustainable manner, countries are scaling up the CCS technologies through a series of initiatives [47]. Such the bench- and laboratory-

scale system is an essential early-stage tool to evaluate and scale novel technologies, which is highly automated and customized and is a precursor to pilot- and demonstration-scale plants. The pilot plant is a pre-commercial system for novel technologies. The purpose is to learn about novel technologies and then apply the gained knowledge to the design and research of the larger demonstration plant and further clarify research objectives and financial support. Demonstration-scale plants differ from pilot-scale plants in that their equipment and processes are more akin to commercial scale operations and can produce large volumes of final products for market testing, deciding whether to commercialize. Commercial scale refers to the scale of product quantity and the process that meets the expected technical economy and scale of production, which is sufficient to support the anticipated supply demand of the product. Figure 2-1 summarises the technology readiness level (TRL) of CCS technologies. A large number of CCS technologies is stuck at TRL 3, 6 and 7, which need further investment to put forward.

Table 2-1 Carbon neutral goals by country[48]

Country/region	Progress	Date to achieve carbon neutral
Bhutan	Achieved	Negative emissions began in 2018
Suriname	Achieved	Negative emissions began in 2014
Uruguay	Under discussion	2030
Finland	Policy document	2035
Austria, Iceland	Policy document	2040
Germany	Policy document	2045
Sweden	Law	2045
UK, France, Denmark, Hungary, and New Zealand	Law	2050
European Union, Spain, Chile, South Korea, and Canada	Proposed legislation	2050
Japan, US	Policy document	2050
China	Policy document	2060

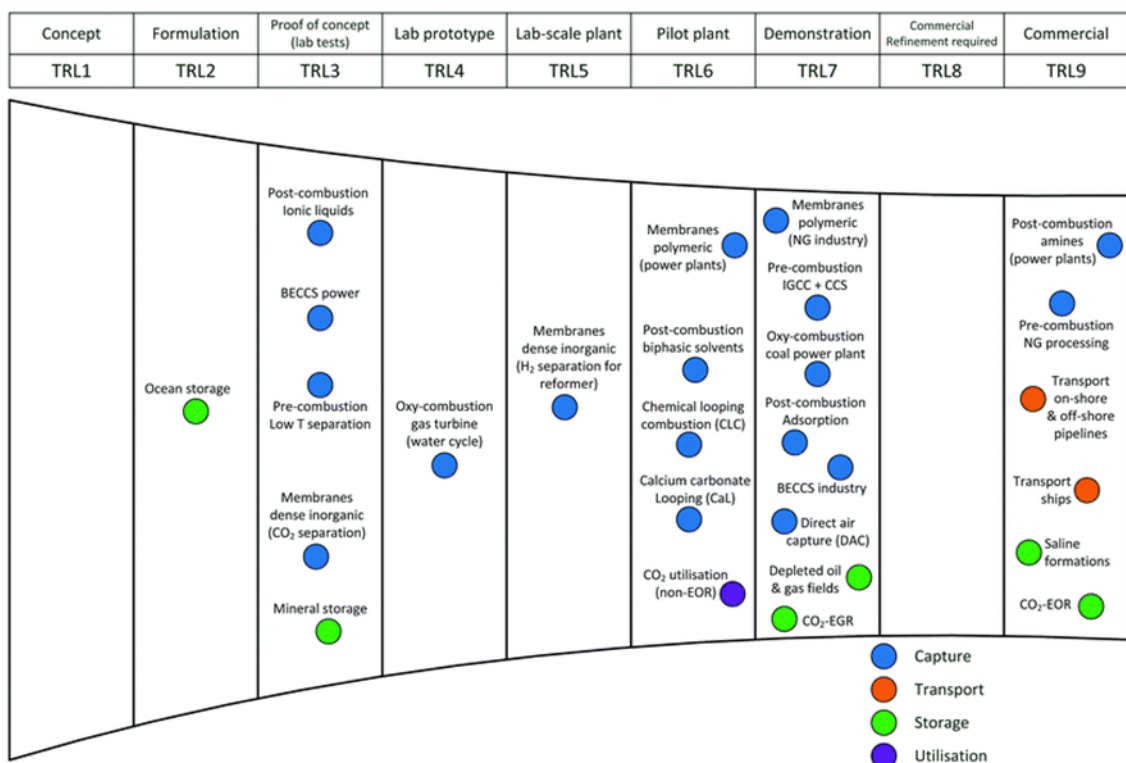


Figure 2-1 Current technology readiness level of carbon capture, storage and utilisation technologies [49]

2.1.1 Pre-combustion capture

Pre-combustion capture [50] refers to the separation of carbon-containing components from the fuel before combustion, mainly used in the Integrated Gasification Combined Cycle (IGCC) power station. It is considered one of the most promising routes of CCS technology in the future. In conventional IGCC power plants (Figure 2-2), coal is gasified to syngas (H_2 , CO and CO_2) by steam and oxygen at high temperature and pressure. And then, the CO_2 is separated, obtaining relatively clean hydrogen-rich fuel gas. Then, the remaining gas is utilised to drive a gas turbine to generate power. The recovered heat is utilised to generate steam and drive a steam turbine to generate power.

In Figure 2-2, the IGCC power station introduces the water-gas shift (WGS) unit, applying carbon capture technology before combustion. The carbon monoxide

(CO) in gas reacts with water vapour (H_2O) to form CO_2 and hydrogen (H_2) and then separate the CO_2 . The syngas shifts in the WGS reaction generating additional H_2 , and CO can be converted to CO_2 . In this case, CO_2 is captured from the converted syngas, while the remaining hydrogen (H_2) is burned in a gas turbine producing electricity. Pre-combustion capture could avoid the disadvantages of large gas flow and low CO_2 concentration compared with post-combustion capture in conventional CFPPs. For IGCC carbon capture power station, what kind of CO_2 separation technology should be adopted, how to select and match other unit technologies, and what is the optimal range of carbon capture rate are all critical issues that need to be studied and paid attention to.

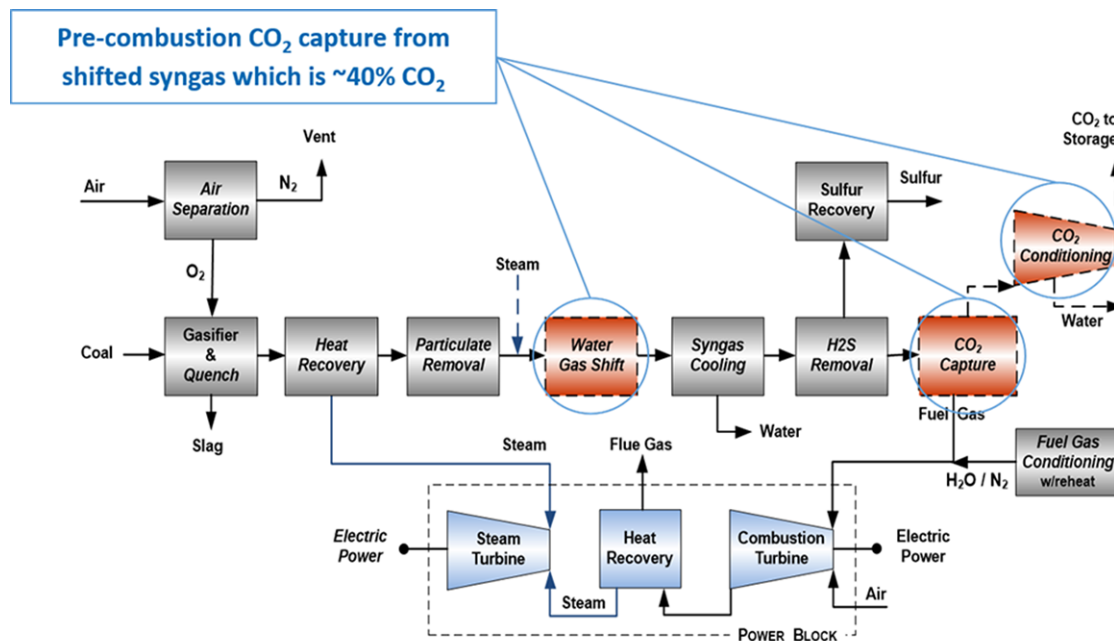


Figure 2-2 Process diagram of an IGCC power plant integrated with CCS [51]

Currently, the research and development of CO_2 separation technologies for pre-combustion capture technology [52] are mainly sorted by advanced solvents, adsorbents and membrane technologies, with a special focus on high-temperature novel materials, process improvement and nanomaterials. In addition, hybrid technology combining the attributes of multiple technologies are investigated.

Solvents [53] absorbing CO₂ involve chemical or physical processes from syngas into liquid carriers. Then it is able to be regenerated with higher temperature and lower pressure. Aiming to improve the efficiency of IGCC plants, current research topics include recovering CO₂ at higher regeneration pressures, improving selectivity to reduce H₂ losses, and improving the CO₂ carrying capacity of the solvent at higher temperatures.

Adsorbent technology [54] is being developed to improve the techno-economic performance for CO₂ separation in IGCC power stations. The research and development objectives are to maintain the high adsorption loading capacity of adsorbent, reduce the attrition loss of adsorbent in multiple regeneration cycles, and perform efficiently under high-temperature conditions in the IGCC system, so as to avoid the required cooling and reheating of syngas. The adsorbent has an optimal and moderate temperature. The temperature of the adsorbent is reduced to meet its required temperature. Also, it preheats the syngas injecting into the gas turbine. The required cooling and reheating of syngas refer to reduce syngas temperature to meet the optimal and moderate temperature of adsorbent, and reheat the syngas, a preheat process, injected into the gas turbine.

Membrane [55] technology is being developed to separate CO₂ and H₂ in produced syngas. The membrane design includes materials of metal, polymer, or ceramic that operate at high temperatures and the separation by various chemical and/or physical processes. The research and development objectives are that the membrane under the condition of the low-pressure drop with high permeability and selectivity, the tolerance for pollutants such as sulphur, and the ability to operate under the system temperature of 260°C.

Novel concepts are being explored, including hybrid systems that combine multiple technical attributes, new process conditions (for example, systems operating at sub-ambient temperatures) and nanomaterials (for example, Nano absorbents [56]). For example, temperature-swing and pressure-swing technologies for regeneration are combined to improve techno-economic performance, and CO₂

capture is directly applied with WGS reactions for balancing the production of CO₂ and H₂ and eliminating a step of syngas cooling.

Advantages of pre-combustion capture technology include the ability to use a variety of fuels, generated H₂ used as the transportable fuel or other products, potential improvement in system efficiency by increasing the technology integration. Disadvantages are higher capital cost, the reliability of all components of integration and reduced short-term flexibility.

2.1.2 Oxy-fuel combustion capture

According to the technical-economic comparison and the development trend of oxy-fuel combustion technology, it is one of the most prominent representatives of CCS technology in CFPPs [57] (Figure 2-3). From a technical point of view, the oxy-fuel combustion technology is a system that the fuel is combusted in an oxygen-rich environment, which is the mixture of O₂ (95vol%) from ASU and partially recirculated flue gas (around 70%) to replace air, and thus the concentration of CO₂ in the exhaust flue gas is increased. The recycled flue gas is utilised to control the temperature of the internal combustion chamber and make up the volume of the missing N₂ to carry enough heat to ensure heat transfer and boiler efficiency [58]. The main components of the flue gas are CO₂ and water vapour by virtue to the fuel combusted in the oxygen-rich and nitrogen-lean environment. It also includes small concentration impurities such as argon and oxygen. After that, the stream is condensed for removing H₂O and is compressed for permanent storage or utilisation.

In such oxy-fuel combustion technology, the CO₂ can be captured through technologies, such as cryogenic separation and distillation process. Relevant studies show that the purity of CO₂ in dry flue gas is able to reach 95%, which is beneficial to the purification, storage and utilization of CO₂. The technology has several other advantages. For example, high combustion efficiency, low NO_x emission, low flue gas loss and low flue gas volume [59,60]. Moreover, it presents

an excellent performance in integrated emissions control by reducing flue gas flow. Finally, the unit volume of the oxy-fuel combustor can reduce to 20% of that of the air-fired combustor [61]. Disadvantages, also challenges, [62] are the higher energy penalty from air separation unit and CO₂ compression and purification unit, higher capital cost (typical to all CCS technologies), the lack of evidence from large-scale plants with the various operating condition, and risk of CO₂ leakage.

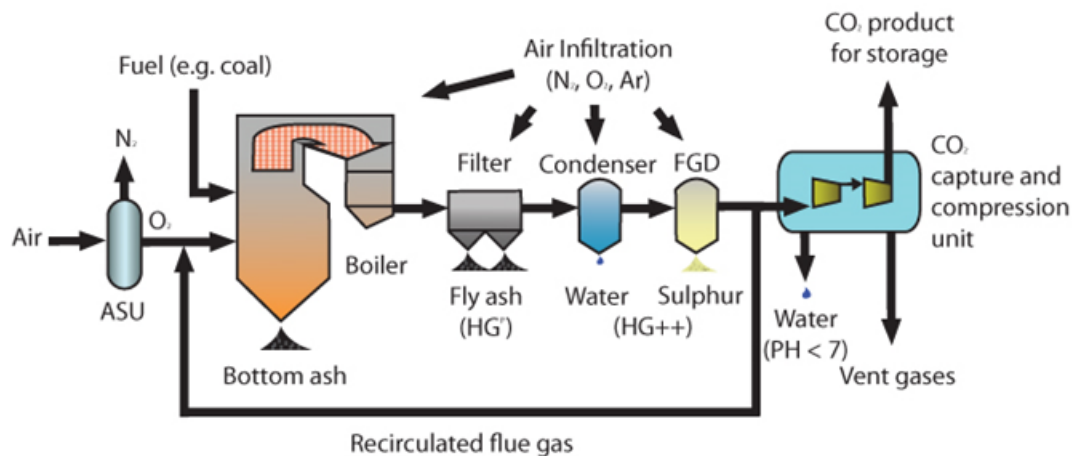


Figure 2-3 Oxy-fuel Combustion Process diagram [61]

2.1.3 Post-combustion capture

Post-combustion capture technology (Figure 2-4) captures CO₂ from the exhaust flue gas after the fossil fuel has been burned. Its most significant challenge is to separate CO₂ from the flue gas containing large amounts of N₂. The research and development chiefly focus on advanced solvents, solid sorbents, and membrane systems. Such solvents and membranes of hybrid technologies are investigated.

Solvent-based CO₂ capture [63] is based on the direct contact of CO₂ and the liquid solvent by chemical or physical absorption. The solvent is then regenerated for discharging CO₂ by increasing its temperature and/or reducing its pressure. The high-level CO₂ capture can be achieved, but it faces a significant energy penalty for solvent regeneration. The research and development of post-combustion

include advanced solvents with lower regeneration energy and selectivity for the flue gas.

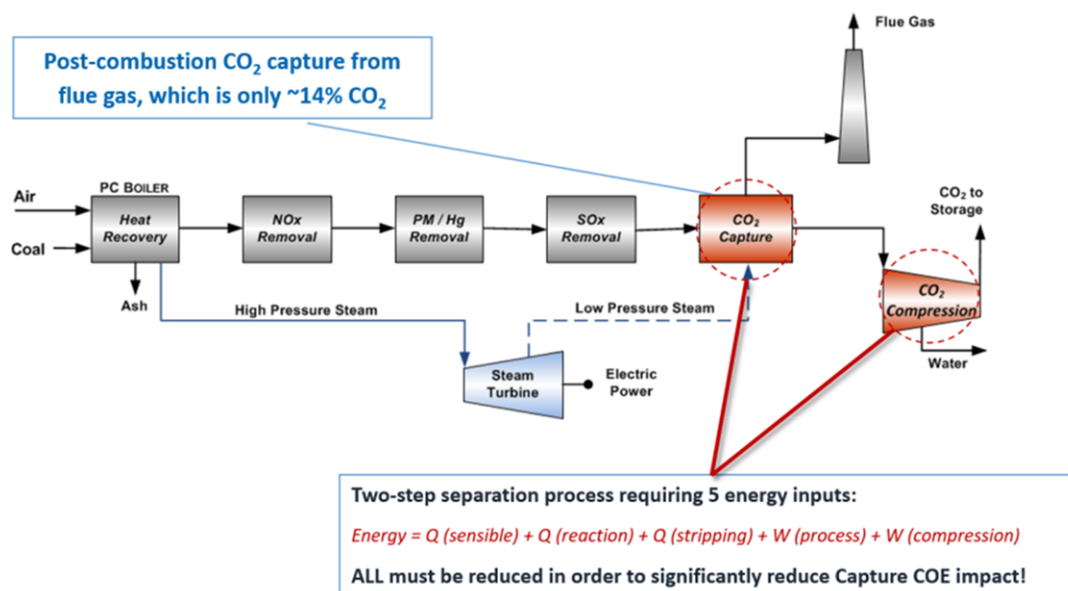


Figure 2-4 Post-combustion in a pulverized coal-fired power station [64]

Sorbent [65] potentially has advantages over the aqueous solvent by virtue to that the heat capacity of solids is significantly lower than that of water. The research and development of sorbents aim to produce the low-cost sustained sorbent with high selectivity and absorption capacity for CO₂ and high-performance under cycling operation.

Permeable or semi-permeable membranes [66] use CO₂ capture for selective transport and separation of CO₂. Its research and development chiefly require that the membrane should have properties of the low-cost, sustained, higher-permeability, high-selectivity, higher stability on thermotics and physics, and high resistance to contaminants in the flue gas.

2.1.4 Bioenergy with carbon capture and storage

BECCS, in addition to CCS, is one of the negative emission technologies that has been deemed crucial to achieving the GHGs reduction targets set by IPCC [7]. This

is since, in the future, BECCS is anticipated to stabilizing the rise in global mean temperature at low levels. BECCS is defined as driven by the fact that biomass consumes CO₂ from the air for growing. If CO₂ is then captured and stored after biomass combustion, CO₂ is permanently removed from the atmosphere, allowing BECCS to be distinguished by negative CO₂ emissions. Hence, BECCS's feasibility will profit from the emission trading system (ETS). For example, the EU Emissions Trading System (EU ETS), launched in 2005, was the world's first large greenhouse gas emissions trading system in the world and kept the largest. The system operates under the 'cap and trade' principle. The cap is the upper limit of the GHG emissions that can be emitted by the installations (power plants, industrial factories and aviation sector) and reduces as time goes on, resulting in the decrease of the total amount of GHG emissions. Emissions allowances are able to be bought and received by the installations. If the emissions of an installation exceed its emissions allowances, the installation is able to purchase the allowances from others. Conversely, the installation can sell the allowances to others needed by virtue to its efficient decarbonisation.

By 2019, six CO₂ capture schemes, including ethanol bio-refinery plants and municipal solid waste recycling centres, were in service, and five BECCS projects were scrapped owing to the difficulties of securing planning approval and undesirable economic profitability. The Illinois Basin Decatur Project, the precursor of the Illinois Industrial CCS (IL-ICCS) project, successfully captured and stored 1 MtCO₂ over 3+ years [67]. Other proposed and current BECCS projects, however, are considerably smaller. In early 2019, the Drax Power Station has started operating the innovative BECCS plant with the C-Capture technology possessing a capture capacity of 1 tCO₂/day [68]. Although the amount of current and proposed projects is encouraging, hundreds of such projects are required worldwide if BECCS is to make a meaningful contribution to achieving decarbonisation goals.

The significant challenges of BECCS are the low efficiency and high LCOE resulting from the lower energy density and higher biomass price compared to that

of coal, which obstructs the implementation of BECCS. In addition, the deployment of BECCS is restricted by the supply of sustainable biomass, CO₂ storage capacity and future technical performance of biomass conversion and CCS technologies [69]. Expansion of sustainable bioenergy production is required under strict emission conditions. Intensive production of bioenergy can place severe pressure on natural resources, particularly land and water, as well as competition among food, feed and energy sources. Thus, it potentially results in contentious economic, ethical and environmental issues. Al-Qayim et al. [70] has reported that it is more commercially viable to add oxy-fuel combustion to BECCS than to apply post-combustion CO₂ capture. Their analysis found that the efficiency of the wood pellet-fired oxy-fuel combustion power plant was 3.8 percentage points higher than that for the same power plant with post-combustion CO₂ capture (26.4 percent). In comparison, the LCOE of the oxy-fuel combustion case (~160 €/MW_{el,h}) was 6% lower. This figure, however, was almost double that for the coal-fired oxy-fuel combustion (~86 €/MW_{el,h}).

2.2 Oxy-fuel combustion system for coal-fired power plants

2.2.1 System composition

This section introduced five major systems [71] for an oxy-fuel power plant with CCS: ASU; oxy-fuel steam generator island; steam turbine cycle island; CO₂ purification unit (CPU); balance of plant (BOP).

2.2.1.1 Air separation unit

The ASU produces nearly pure O₂ for the oxy-combustion steam generator island. The cryogenic ASU is the only mature technology for an oxy-fuel combustion power plant, including raw water filtration, air compressors, water vapour and trace CO₂ removal, pre-refrigeration, and the cryogenic cold box and an expansion turbine. There are four differences of ASU requirements in between the power plant and industry: a) multiple ASU trains are required in an 800 MW_{el, gross} oxy-fuel combustion power plant due to the O₂ consumption (20,000 tonnes per day) is

significantly higher than the built today; b) 95%-97% of O₂ purity is sufficient for an oxy-fuel power plant, rather than 99.5% is required in the industry. The expensive oxygen-argon separation can be saved when the minimum requirement of O₂ purity is 97%; c) the O₂ delivery pressure to oxy-combustor is 2 bar in the oxy-fuel combustion power plant, which is generally lower than that in the industry; d) the ASU plant in the oxy-fuel combustion power plant is required have sufficient flexibility in responding to load-following dispatch. Designers could reference the above differences to make improvements in reducing the energy penalty and costs.

2.2.1.2 Oxy-fuel steam generator island

In the oxy-fuel steam generator island, the main components include O₂-flue gas mixing, fuel metering, heat transfer surface of furnace and steam generator, O₂ heater, particulate control device and ash handling, SO_x removal system, NO_x removal system, and recycle fan, induced draft fan and air-fired stack.

2.2.1.3 Steam turbine cycle island

The steam turbine cycle island includes significant components of high-, intermediate- and low-pressure turbines and controls, the condenser, the cooling water circulation and cooling towers, the condensate and Feedwater pumps, and the feed water heaters and deaerator. Due to the identical steam turbine system in both air- and oxy-fuel combustion, the required materials and processes for oxy-fuel combustion become available by using the existing air-fuel combustion, enabling the same cycle efficiency. It is different from the post-combustion technology in that the post-combustion need to modify the steam turbine cycle for regenerating the CO₂ capture solvent. The net power output of the oxy-fuel combustion power plant is mainly impacted by the ASU and CPU. The cost of the air- and oxy-combustion power plants should be identical by virtue to them identical components of steam turbine cycle island.

2.2.1.4 CO₂ purification unit

In the CPU, the main components include indirect and direct contact flue gas cooler, compression with inter-cooling, deep drying, active carbon bed, cold box, cold-box vent gas CO₂ recovery, and injection pressure compression/cooling. The CPU has great uncertainty in overall performance and cost due to there is no typical design specification of CO₂ purity delivery to a pipeline or a storage site. The raw flue gas generally includes CO₂ (around 80%), nitrogen and argon. The process with the lowest energy requirement of the CPU is only to cool and dehydrate the raw flue gas and then to compress and inject into the geological storage structure without further purification, which achieved 100% CO₂ capture and the lowest cost for the power plant. However, in the maximum requirement of CPU, if the higher CO₂ purity is required in enhanced oil recovery (EOR), the CO₂ purification process is required by a partial condensation that increases the capital cost and energy penalty of the oxy-fuel power plant [72]. Over 95% CO₂ concentration could be achieved by employing a partial condensation process with a single- or double-stage flash. The CO₂ impurities (uncondensed CO₂) existing in the vent gas of the partial condensation process can be removed by the developing vent gas recovery CO₂ technologies, which is expected to reach over 98% CO₂ capture efficiency.

2.2.1.5 Balance of plant

The balance of plant has similar deployment on the air-combustion power plant, including electric generator, transformer and switchgear, auxiliary power switchgear, water supply and treatment, coal yard and handling, ash handling and disposal, and wastewater treatment and disposal. Moreover, the cost of oxy-fuel combustion can be predicted from the air-combustion power plant with the same capacity.

2.2.2 Background, motivation and existing challenges

For years, CO₂ from natural sources and industrial processes has been extensively used in the oil and gas industry to extract oil from oil fields known as EOR. In some cases, the combustion flue gas has been used in EOR [73]. An idea of oxy-fuel combustion was proposed for achieving a high concentration of CO₂ in 1982 [74]. Partial exhaust gas recycles to the furnace for controlling inner combustion temperature. It was firstly tested in a 3 MW_{th} pilot facility at the University of North Dakota with the partner of Argonne National Laboratory [75]. Oxy-fuel combustion has been used for a long time in several industry sectors, such as the industry of glass, aluminium, cement, steel and incineration, which primarily aims to the higher productivity, lower fuel consumption and lower NO_x emissions. The scale of these applications is smaller than that of power generation and generally without flue gas recycle.

With climate change concerned since the late 1990s, oxy-fuel combustion rapidly developed with the pilot-scale testing to the industrial-scale demonstration introduced in Section 2.2.3. The oxy-fuel system is less complex than the pre-combustion and post-combustion that require an additional process to capture CO₂. In addition, the sections of coal combustion and air separation of oxy-fuel combustion are mature and have been widely operated. Also, it has the dual firing capability (switch between air combustion and oxygen-rich combustion), which is able to meet peak load demand under air firing conditions. Due to the flue gas recycle in oxy-combustion, the NO_x formation is significantly lower than the air combustion, and the flue gas volume is around one-quarter of that in air combustion, which leads to a smaller flue gas control equipment with cost saving. Furthermore, the integration of emission control of SO_x, NO_x and mercury might be acted as part of CO₂ capture [76].

The main challenge for oxy-fuel combustion is the high energy penalty in ASU and CPU with the further cost increase. Although the ASU using cryogenic air separation is improved, it still requires 200kWh generating per ton of O₂, which is

an energy-intensive process in oxy-fuel combustion technology with a further significant efficiency reduction of the entire system. The CPU is a multi-stage compression and cooling system for purifying and compressing the exhaust gas. It aims to produce high purity CO₂, satisfying the criteria of CO₂ transport and storage by removing the water vapour, O₂, N₂ and Ar.

2.2.3 History of and developments in oxy-fuel combustion

The development of new concepts and designs for oxy-fuel combustion technologies represents an emerging opportunity in fossil fuel conversion technologies. It aims to improve the efficiency of the combustion process and reduce the environmental impact by capturing CO₂ emissions through CCS [61].

Since the 1990s, comprehensive research and development of oxy-fuel combustion CO₂ capture have been conducted. Laboratory-scale tests focus on fundamental studies, such as ignition characteristics, flame stability, rate of char combustion, boiler performance [77] and flue gas composition [78], heat transfer behaviours and retrofit of the existing boiler. The pilot-scale plant has been operated worldwide, such as Canada, the USA, Japan and Europe. Based on the previous operation experience, it is proved that the furnaces are able to retrofit from air-combustion to oxy-combustion with lower air infiltration, high CO₂ concentration flue gas and significantly lower NO_x emissions.

Several major pilot plants and pre-commercial demonstration projects (Table 2-2) lowering than the capacity of 100 MW_{th} have been demonstrated worldwide to prove the technical feasibility of oxy-fuel combustion technology, such as those facilities with 15 MW_{th} in the USA [79], 30 MW_{th} in the USA [80], Germany [81], France and Australia [82], 40 MW_{th} in the United Kingdom [83]. The CanmetENERGY established a 0.3 MW oxygen-enriched combustion test system to study flame, heat transfer and pollutant emission of pulverized coal combustion in 1994. IHI Corporation uses oxy-fuel combustion technology in a 100 MW demonstration power plant, and the system also uses the flue gas heat exchanger.

The test results show that denitrification and desulfurization efficiency reaches 70% and 90%, respectively, and the land area is only about 50% of the original one. ALSTOM Power, Inc., in collaboration with US Power Plant Laboratories, the project "control of GHG from pure oxygen combustion in circulating fluidized bed (CFB) boilers" was carried out. Japan's Hokkaido institute of industry (HNIRI) has conducted an international cooperation project on "advanced coal combustion and emission control technology" to study the oxy-fuel combustion mechanism and the emission characteristic of SO₂/NO_x of the bubbling fluidized bed (BFB). A series of experimental studies on oxy-fuel combustion in the CFB has been carried out in Zhejiang University and North China Electric Power University. The Callide Oxyfuel project demonstrated that CCS technology can be applied to CFPPs, generating electricity with virtually no emissions. It became the world's first retrofitted industrial-scale demonstration of oxy-fuel combustion, which was available to be applied to commercial-scale capture-ready power plants and could be scaled to above 350 MW_{el} [84]. CanmetENERGY (formerly CANMET) is currently working on hydroxy-fuel combustion (the 3rd generation oxy-fuel system) in which fuel is burnt in a high-purity O₂ environment without flue gas recirculation, using water or steam as a temperature moderator [85]. The advantage of this configuration is that it reduces capital costs by reducing the mass flow of gas in the system. Nevertheless, novel turbomachinery needs to be developed and driven by steam/gas mixtures as a working medium. Oxy-fuel combustion projects at CanmetENERGY enable industrial customers and stakeholders to jointly develop and review advanced process configurations and design principles of new equipment for the next generation of oxygen-enriched combustion technology. Their achievements have led to industry innovation in this field and guided the pre-commercial demonstration and commercial operation in cost-effective ways [61].

Table 2-2 Major demonstration plants of oxy-fuel combustion [71]

Country	Project name	Fuel	Scale	Type	Start date
Germany	Vattenfall	Coal	10 MWe	New	2008
USA	Babcock& Wilcox	Coal	10 MWe	Retro	2008
UK	Doosan Babcock	Coal	13 MWe	New	2008

USA	Doosan Babcock	Coal	30 MWe	N/A	2008
Germany	Schwartz Pumpe	Coal	30 MWth	New	2008
France	TOTAL	NG	10 MWe	Retro	2009
USA	Jupiter Pearl plant	Coal	22 MWe	Retro	2009
Italy	ENEL Oxy-fuel CCS2	Coal	16 MWe	New	2012
Australia	CSEnergy-Callide	Coal	30 MWe	Retro	2012
Spain	CIUDEN	Coal	30 MWe	New	2012
USA	Jamestown/Praxair	Coal	50 MWe	New	2013
China	Yingcheng	Coal	35 MWth	Retro	2015
USA	Babcock& Wilcox	Coal	100 MWe	New	2015
Youngdong	South Korea	Coal	100 MWe	Retro	2016

Large amounts of oxy-fuel combustion models are established by the digital computing platform, and further related assessment and parameter optimization of the oxy-fuel CFPP are able to provide a dependable reference for industrial application with further cost savings. Regarding the modelling of oxy-fuel combustion and its related assessments, Xiangchun Chen used Life Cycle Assessment to analyse the process of 300 MW CFPP generating electricity for 1 hour under an oxygen-rich atmosphere and calculated the proportion of power consumption of air separation, liquefied recovery of CO₂ and pulverized coal transportation. The Chalmers University of Technology assessed the system's efficiency when converting an 865 MW CFPP into an oxygen-rich combustion plant, which reduced the power supply by 9.1% [86]. IHI Corporation [87], ALSTOM [88], Argonne National Laboratory, CanmetENERGY [88], and Huazhong University of Science and Technology all conducted a techno-economic evaluation on the pulverized coal oxy-fuel combustion system, which concludes that if the conventional boiler is transformed into an oxy-fuel combustion boiler, the net efficiency of the power plant will be reduced by 8-12%, and the power supply with the cost increased by 40-50%. In the evaluation from Xiangchun Chen and Chalmers University of Technology, the power consumption of ASU is calculated by the energy consumption per unit of oxygen production and oxygen supply provided by the ASU manufacturer [89,90]. For the oxy-fuel combustion system,

except for the concentration of oxygen required ($> 95\%$), other state parameters are not required to be high, only need to be in a gaseous state. So, it is necessary to analyse the ASU deeply. As the concentration of oxygen supply increases, the energy consumption per unit of oxygen production increases, while the amount of non-condensable gases in flue gas decreases, thus reducing the energy consumption of flue gas compression and purification units. Therefore, ASU and the flue gas compression and purification units could be coordinated and optimized. Since the CPU condenses and compresses the flue gas, much heat is created so that the heat is able to be comprehensively used to heat the feed water into the boiler, thereby improving the efficiency of the unit.

The simulation works for oxy-fuel combustion also have been down in the literature. Andreas Rodewald and Alfons Kather simulated the oxy-fuel combustion plant system through Aspen PlusTM with its feasibility analysis in thermodynamics and economy [91]. Jongsup Hong simulated the oxy-fuel combustion power station system in Aspen PlusTM and THERMOFLEX[®]. The techno-economic sensitivity analysis was conducted, including combustion temperature in the chamber, the outlet temperature of the boiler system, oxygen purity and O₂ concentration of exhaust gases [92]. Jie Xiong et al. used Aspen PlusTM to simulate the 800 MW ultra-critical oxy-fuel combustion power plant. The results showed that the CO₂ concentration in the flue gas exceeded 80%. The effects of the recirculation rate of exhaust gases and the O₂ concentration at the ASU outlet are also analysed [93]. Jie Xiong et al. conducted the exergy analysis on a 600 MW ultra-critical oxy-fuel combustion power station and concluded that oxygen-enriched combustion boiler has 0.8% higher exergy efficiency than the conventional boiler [94]. Kakaras et al. conducted a techno-economic evaluation for a system of the oxy-fuel combustion power plant fuelling lignite, believing that the implementation of the oxy-fuel technology reduces the net electrical efficiency of the power station by 8.5% [95]. Sivaji Seepana and Sreenivas Jayanti improved the system of the common oxy-fuel combustion power plant by changing the way of recirculating exhaust gases

treatment, and made a detailed thermodynamic analysis of the improved system [96]. Regarding the optimization of the oxy-fuel combustion thermal system, Hongbing Kong used Aspen PlusTM to establish the oxy-fuel combustion with optimization analysis. However, he did not optimize the ASU, and only recovered the heat of the CPU from flue gas, ignoring the large amount of waste heat generated in the ASU. Cormos [97] analysed the performance of coal, lignite and sawdust fired oxy-fuel combustion power plants. Their analysis found that, compared to the reference CFPP without CCS, the considered process would result in efficiency penalties of 9-12% points. The main obstacle of oxy-fuel combustion is a high efficiency penalty arising from the high-purity O₂ output within the ASU (50-60%) and CO₂ compression within the CPU (30-40%), which together account for approximately 90% of the energy penalty [98,99], which has contributed to a rise of 50–95% in the LCOE. Singh et al. [99] contrasted the techno-economic performance of amine scrubbing and oxy-fuel combustion retrofitted by an existing CFPP. Both processes were shown to increase electricity prices by up to 20–30% compared with the reference CFPP without CCS. However, oxy-fuel combustion was shown to be more attractive owing to its lower CO₂ emissions and lower cost of CO₂ capture. Overall, it is stated in the literature that the LCOE for the oxy-coal combustion is between 70 €/MW_{el}h [97] and 106.3 €/MW_{el}h [100], and using sawdust is 78.87 €/MW_{el}h [97]. So, it is necessary to study solutions to reduce energy loss and cost in order to improve the feasibility and large-scale application of oxy-fuel combustion. In order to solve the issue of high-energy requirements from cryogenic-based ASU, Zhou et al. [101] found an integrated chemical looping air separation unit (ICLAS). Their research proved that ICLAS significantly improved the techno-economic performance of oxy-fuel combustion power plants utilising steam and recycled flue gas and demonstrated that implementation would minimise the efficiency penalty of oxy-fuel combustion from 9.5% to 5% and can decrease the LCOE by up to 20%.

At present, the next-generation oxy-fuel combustion technology is investigated at CanmetENERGY, including the near-zero-emission oxy-fuel gas turbine, and the oxy-fuel process and power plant modelling. Experimental studies on the oxy-fuel combustion process of potential fuels, and the development of a comprehensive process for hydrogen and oxygen production, backed by strong research teams and the most advanced pilot scale vertical combustion research facility [61].

2.2.4 Uncertainty for performance and cost

Due to the additional component considered in the oxy-fuel combustion power station, the performance and cost are affected by its uncertainty which is referred to in the following details. The ASU and CPU of an oxy-fuel combustion power station need to be scaled up to a required capacity, which is significantly higher than that in the industry built to date. In addition, the uncertainties are the uncertain specification on the O₂ product purity and CO₂ product purity, respectively, at the outlet of ASU and the outlet of CPU. If needed to meet the emissions standard, flue-gas desulfurization (FGD) and selective catalytic reduction/selective non-catalytic reduction (SCR/SNCR) are required. The last uncertainty is the performance of large-sized CPU, including CO₂ purification systems, partial condensation and automatic refrigeration.

2.2.5 Comparison among oxy-fuel combustion with other primary CCS technologies

In Table 2-3, four cases were introduced, including a supercritical CFPP without CCS (B12A), a supercritical post-combustion CFPP (B12B), oxy-fuel combustion power plant (B1B), IGCC with CCS (Case 2). Detailed information and overall performance of the related cases were listed, including net electrical efficiency, specific CO₂ emission, LCOE and CO₂ avoided cost (AC). The cases were investigated in two reports from the National Energy Technology Laboratory in the United States Department Of Energy (USDOE/NETL) [102,103], consisting of three leading CCS technologies – pre-combustion, oxy-fuel combustion and post-

combustion. From Table 2-3, it can be concluded that the net efficiencies of CCS power plants are around 32% lower than the 40.3% of supercritical CFPP without CCS. The LCOE of oxy-fuel combustion is significantly lower than the pre-combustion, but higher than the post-combustion. This results in a similar trend in the cost of CO₂ avoided, corresponding to the level over 99% CO₂ emissions purity. Confirming purity of the CO₂ product for different applications is essential in reducing the techno-economic performance of the plant due to further purifying the CO₂ by handling the impurities required incrementally higher cost.

2.3 Chemical looping combustion system

The emerging chemical looping combustion (CLC) technology is, in essence, a variety of oxy-fuel combustion technology to investigate its potential for reducing energy penalty and cost compared with conventional oxy-fuel combustion.

2.3.1 Chemical looping combustion with solid fuels

In CLC, the fuel directly combusts with oxygen carriers (OCs), avoiding the direct contact between the fuel and air, to produce a flue gas stream from the fuel reactor (FR) primarily containing CO₂ and H₂O while generating heat for electricity production. The flue gas stream from the FR is treated to a nearly pure CO₂ stream by condensing out the steam, avoiding utilising an energy-intensive ASU [104].

Table 2-3 Techno-economic performance of CCS technologies for CFPP

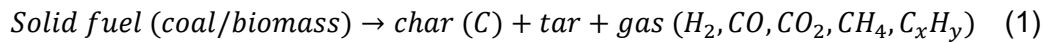
Source		USDOE/NETL			
CO ₂ capture scenario	No capture	Post-combustion capture	Pre-combustion capture	Oxy-fuel combustion capture	
Technology	Supercritical pulverized coal	Cansolv	IGCC-Selexol	Advanced Cryogenic ASU	
Case	B12A [102]	B12B [102]	B1B [102]	Case 2 [103]	
Steam condition	24.1 MPa/593°C/59 3°C	24.1 MPa/593°C/59 3°C	12.4 MPa/533°C/53 3°C	24.2 MPa/593°C/62 1°C	
Gross power	685	770	696	747	
Auxiliary Power (MW)	35	120	177	197	
Net power (MW)	650	650	519	550	
Net efficiency (% _{HHV})	40.3	31.5	32.5	31.5	
Specific CO ₂ emission (kg/MWh)	777.5	99.3	97.5	77.4	
CO ₂ emissions purity (mol%)	12.46	99.95	99.03	99.99	
CO ₂ capture (%)	0	90	90	92.2	
Total plant cost (\$/kW)	2099 ^a	3800 ^a	6209 ^a	3484 ^b	
LCOE (\$/MWh)	64.4 ^a	114.3 ^a	175.0 ^a	136.4 ^b	
AC (\$/tonne)	0	73.5 ^a	162.7 ^a	113.4 ^b	

IGCC: Integrated Gasification Combined Cycle

a: base year of 2018; b: base year of 2011

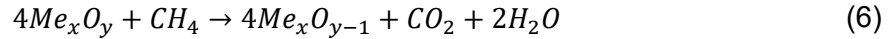
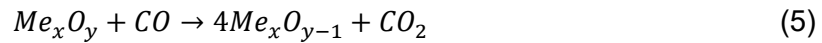
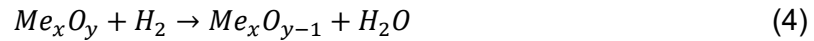
Most researchers paid more attention to chemical looping with gaseous fuel, but solid fuel was adopted as well, which has been popular in the past several years. The earliest study of CLC with solid fuel using copper (Cu) and iron (Fe) as OCs appeared in 1951 [105]. Half-century later, researches of the same OCs was

applied in the new studies [106,107]. Leion et al. studied different fuels and OCs in a small laboratory fluidized bed [108–110]. What has followed were the comprehensive review papers regarding the overall CLC [111], the solid fuel [112] and its reactors [113], which showed great potential for CLC with solid fuels. CLC for solid fuels is also referred to as in-situ gasification chemical looping combustion (iG-CLC) due to its complicated reaction process comparing to CLC for gaseous or liquid fuels. Reactions of CLC for solid fuels are a two-step process—devolatilization of the fuel and gasification of the char. Solid fuel particles are devolatilized within seconds, and then the char is gasified by steam/CO₂ taking several minutes. The char gasification kinetics referenced the sources from Johnson [114] and Fennell and Anthony [115]. Finally, the gaseous products react with OC generating CO₂ and H₂O. The principal reactions of converting the solid fuel to gas include:



In the experiments from Zeng et al. [116], the coal char located upstream of OC because the coal char and OC were packed into the fixed bed separately. Therefore, the OCs were reduced by a gaseous product from gasification. Due to the direct mixing of OCs and char, particle conversion can significantly enhance the reaction rate of char conversion. In addition, higher char conversion can achieve by adjusting the concentrations of gasification enhancers (CO₂ or H₂O). However, excessive gasification enhancers can result in oxygen carrier oxidation, which must be carefully done.

The devolatilized and gasified gas products, such as H₂, CO and different hydrocarbon, react with the metal oxide (Me_xO_y) through the below reactions. The OCs reaction kinetics referenced the sources from Abad [117] and Fennell and Anthony [115]. The CH₄ refers to all hydrocarbon from devolatilization.



2.3.2 Comparison between coal and biomass-fuelled chemical looping combustion

Coal and biomass are two representative solid fuels applied to CLC. Much of the early research focused on coal which has been extensively used. Fuelling low-rank coals such as bituminous (e.g. Illinois No.6 coal) achieves higher CO₂ capture efficiency and fuel conversion rate [118] by virtue to its rich volatiles in hydrocarbon compared to the high-rank coals and the CO₂ from volatiles is, in theory, completely captured in FR. So choosing an OC with high reactivity toward hydrocarbon is essential. Such as a 0.5 kW_{th} CLC plant in Institute of Carbochemistry in Spanish National Research Council (ICB-CSIC) proves that with publications from Adánez et al. [119] and Cuadrat et al. [120]. The higher rank coals, such as anthracite, have a slow gasification rate. A stripper for improving the CO₂ efficiency is required to separate unreacted char and send them back to FR.

In the last few years, there is a growing interest in biomass-fuelled CLC due to the concept of BECCS [121]. Also, related studies have evaluated the different properties between biomass and coal impacting CLC. In addition, biomass reacts more quickly than low-rank coal because, on a dry basis, the higher volatile content of biomass is around 75%. After devolatilizing the remaining char, it is porous and contains several active sites and gasification catalysts [122]. In general, the ash content is low, and the moisture content varies widely. The apparent advantages of using biomass are generally because of higher reactivity than coals. An interesting advantage of biomass-fired CLC is that the fuel reactor does not have any heat-transfer surfaces. Fuel is combusted in FR, extracting heat from the air reactor (AR) through a highly exothermic reaction between air and OC. Such as in a conventional biomass-fired CFB power plant, in general, high content of alkali [123]

exists in the biomass, resulting in superheater corrosion and formation of deposits and further leads to a limitation of steam temperature to 500-540°C. However, the chemical looping CFB power plant is not restricted, and the steam temperature could be higher than 600°C, which benefits net electricity efficiency.

2.3.3 Oxygen carriers

Regarding the OCs, from a commercialization standpoint, the desired OC should have the following characteristics: 1) sufficient reactivity with fuel and oxygen during the oxidation reaction and reduction reaction in the reactors, and complete conversion with the fuel to CO₂ and H₂O; 2) high resistance to attrition and fragmentation to minimise its losses; 3) no presence of agglomeration; 4) low cost; 5) environmentally friendly, low risk for health and safety; 6) adequate O₂ transfer capacity. Much of OC research work has chiefly studied Me_xO_y based on iron, nickel and manganese. Also, the CaSO₄ as the OC has been tested. Similarly, they were investigated in Section 5.

Iron-based OCs have low prices due to they can be achieved from ores and industrial residue. The oxidized form is Ferric oxide (Fe₂O₃), with its reduction form is ferroferric oxide (Fe₃O₄). Of the Fe₂O₃, Fe atoms have an oxidation state of +3. It has a melting point of 1565°C and a density of 5.24 g/cm³. Fe₃O₄ is a kind of mixed valence oxide of iron composed of ferrous oxide and iron oxide (FeO·Fe₂O₃). Of these, two Fe atoms have an oxidation state of +3 and one of +2. It has a melting point of 1597°C and a density of 5.18g/cm³. Due to its thermodynamics, reducing to lower oxidation states (ferrous oxide (FeO) or even Fe) is not possible under the condition of complete fuel conversion. Manufactured iron-based OCs generally have dilatory reactivity with CH₄, rather than much better reactivity towards syngas components, i.e. H₂ and CO [124]. These metal oxides are non-toxic and possess non-agglomeration characteristics at higher operating temperatures [125]. The Fe₂O₃ oxygen carrier, with its properties in this paper, is assumed to be a manufactured material, same as the reference case, which is a comparatively expensive fabricated material ranging from 2,000 to 10,000 € per ton (Table 5-3).

In addition, Fe_2O_3 -based OCs have cheaper forms, such as a natural ore (e.g., hematite, ilmenite) or a waste material (e.g., red mud).

Manganese-based OCs, such as manganese ore, usually have Mn_3O_4 as an oxidized form. MnO is a reduced form that cannot be further reduced under any reduction conditions [126]. Mn_2O_3 was easily reduced to MnO under combustion conditions, which mostly could be oxidized to Mn_3O_4 . In the literatures, there are relatively few experimental data on Manganese-based OCs and the data are not comprehensive. In this study, a kind of low-cost manganese ore [127] was selected tested for CLC with solid fuels, mainly constituted by Mn_2O_3 and Fe_2O_3 with higher than 7 wt% of oxygen transport capacity representing the oxygen-carrying capacity of the OC (Table 5-3). Furthermore, it has higher combustion efficiency and a higher char gasification rate compared to the ilmenite. For one thing, higher combustion efficiency is associated with its higher reaction activity to H_2 and CO [128]. For another thing, the higher char gasification rate is related, both with its higher reaction activity to syngas and the catalytic components, Na and K, in manganese ore [129]. However, a weakness of the manganese ore is its high attrition rate that decreases its lifetime in operation [130], which possibly improves by heat-treating the particles. Surprisingly, even manganese ores have higher reactivity than iron ores and ilmenite, but CLC operational with manganese ores is smaller than that with ilmenite or iron ores which could be because the manganese ores are less easy to access.

Ni-based OCs, in the limited experiment, showed high reactivity with methane rather than solid fuel. Furthermore, Nickel oxide did not show good overall performance due to its toxicity, expensive cost and deactivation with high sulphur content [131,132]. The cost of handling the fly ash and bottom ash could be increased if the bed material is harmful to the environment. NiO supported By Al_2O_3 was first investigated in the 1990s [133] and later in Chalmers.

Afterwards, it was successfully operated and validated within the long-term operation in a European Union (EU) project GRACE (Grangemouth Advanced CO_2

Capture) and CLC Gas Power [134]. Comparing Ni-based OCs to other OCs in several studies, it is more suitable for gaseous fuel.

CaSO_4 as OC have high oxygen transport capacity ($R_{\text{OC}}=47$ wt%) in contrast to Fe and Mn-based OCs, which is beneficial to the carbon capture rate resulting from the lower OC circulation rate between fuel reactor and air reactor. However, problems were founded regarding the lower reactivity of anhydrite ore and sulphur release conversing CaSO_4 to CaS . The lower reactivity reflects in thermodynamic constrain with limitation of CO and H_2 conversion [115]. The sulphur release is hard to be analysed due to varied impact factors, such as reactors temperature, the sulphur content of fuel and degree of fuel conversion [115]. A 3 MW_{th} CLC unit designed by Alstom implemented CaSO_4 as an OC, adding limestone for avoiding sulphur emission. In addition, limestone is more reactive to convert to CaSO_4 than anhydrite ore.

2.3.4 Chemical looping combustion with gas agent CO_2 and H_2O

H_2O and/or CO_2 were served for the gasifying agent and fluidizing agent, which is considered in the iG-CLC, rather than the chemical looping with oxygen uncoupling (CLOU) process does not require a gasification agent. Since the slow solid fuel gasification process (gasifying agent required) is avoided in CLOU, the solid fuel can react directly with the gas phase oxygen. By increasing the mole ratio of H_2O to fixed Carbon (C_{fix}) from 1.2-1.8, the net electric efficiency of iG-CLC decreases 1.3% [135]. A higher H_2O fraction in the flow of $\text{H}_2\text{O}/\text{CO}_2$ gasifying agent could increase the CO_2 capture efficiency. Nevertheless, in general, using either H_2O or CO_2 would not significantly impact the process, such as the combustion efficiency in FR and the oxygen demand [136]. Furthermore, recycling part of hot exhaust gases to FR could reduce steam generation and its energy consumption resulting in an efficiency increase, validated in the literature [137,138].

2.3.5 Energy penalty and cost of Chemical looping combustion

The CLC power plants have net efficiency of around 35% for the supercritical steam cycle [135] and around 41% for the ultra-supercritical steam cycle [135]. The energy penalty of CLC is mainly from the CO₂ compression unit. Rather than the typical CCS technologies, the net electricity efficiencies of pre-combustion, post-combustion and oxy-fuel combustion are around 34%, 34-36% and 34-37%, separately, requiring additional energy-intensive steps for gas separation. Compared with a conventional power plant without CO₂ capture, in general, the energy penalty and the CO₂ capture capability for coal-fired CLC power plants is estimated to be 2-3% and 95%, respectively, but 7-9% and 90% for the typical carbon capture technologies.

Cost of capture in chemical looping applications drawn more attention, which was compared with other CCS technologies. LCOE of iG-CLC in the US market is in the range 105-120 \$/MWh, which is around 20 \$/MWh higher than the conventional power plants without CO₂ capture and close to the advanced post-combustion with amine (114.3 \$/MWh) [102] and advanced oxy-fuel combustion (121.5 \$/MWh) [103]. The LCOE of solid-fuelled CLOU in the EU market is between 84 €/MWh to 92 €/MWh, lower than that of other CCS technologies. In addition, the cost of capture on CCS is in the range of 13-67 €/tCO₂ [46]. It was estimated that the CO₂ avoidance cost for post-combustion is 28-37 €/tCO₂ and for oxy-fuel combustion is 30-58 €/tCO₂. A study of CLC compared with CFB combustion, from Lyngfelt and Linderholm [139], CO₂ avoided cost was 12-27 €/tCO₂ with 10 €/tCO₂ from CO₂ compression. Even so, continuing research for its commercialisation is necessary in the future in terms of, for example, various financial structures.

2.4 Supercritical carbon dioxide Brayton cycle

2.4.1 Introduction and background

Due to energy conservation and environmental conservation have attracted widespread attention, electricity demand is enslaved to the energy demand and

supply, and environmental concerns. The improvement on the energy efficiency is a way to tackle the problems, which is able to be achieved by the sCO₂ Brayton cycle due to its compact configuration and high thermal efficiency in the relatively low range of TIT (450-750°C). CFPP has been the primary mode of power generation since the 1700s. Most of the CFPP employed the Rankine cycle converting thermal energy to electrical energy. During the past decades, the steam Rankine cycle has made significant progress of overall performance, including power plants operating at 31.0 MPa and 621°C with around 40%_{HHV} plant efficiency in American Electric Power's Philo Unit 6 [140], operating at 26.2 MPa and 600°C with 40%_{HHV} per unit in American Electric Power's Turk plant [141,142]. It could be envisioned that the plant efficiency and cycle efficiency of the advanced ultra-supercritical power plant operating at 34.4 MPa and 760°C are in the range 43.7-44.1%_{HHV} and 52%, respectively [143]. Assuming similar costs, the thermodynamic efficiency of indirect sCO₂ Brayton cycles will need to achieve the plant efficiency between 50%_{HHV} and 55%_{HHV} to compete with the present and future advanced steam Rankine cycle power plant [39]. Compared to the conventional steam Rankine power cycle, numerous studies have shown that the Brayton cycle using sCO₂ as the working medium have the potential to achieve higher efficiency than the steam Rankine cycle, which will further result in lower fuel cost, water usage and CO₂ emissions. Moreover, the higher-pressure requirement on CO₂ in the cycle results in the high density of sCO₂, resulting in compact equipment sizes and plant area, so lower capital cost.

CO₂ has a critical point when the pressure is 7.37 MPa and temperature is 304.12 K. At the supercritical state, the sCO₂ possess an approximated-liquid density and an approximated-gas viscosity and diffusion, without toxic, corrosive, flammable and explosive and with adequate supply and reasonable price. The properties of sCO₂ dramatically vary approaching its critical point (Figure 2-5 and Figure 2-6), such as apparent variation of the ratio of specific heat approaching to its critical point and the temperature of the peak ratio of specific heat increased by pressure

rise (Figure 2-5). Such feature results in two problems: one is the partial overlap in flow field resulting from the unsmooth property variation near the critical point, one is the two-phase region may appear in the flow field at the inlet of the compressor owing to the impetus of leading edge of the blade. Moreover, supercritical CO₂ has well heat-transfer properties than liquid and gaseous CO₂.

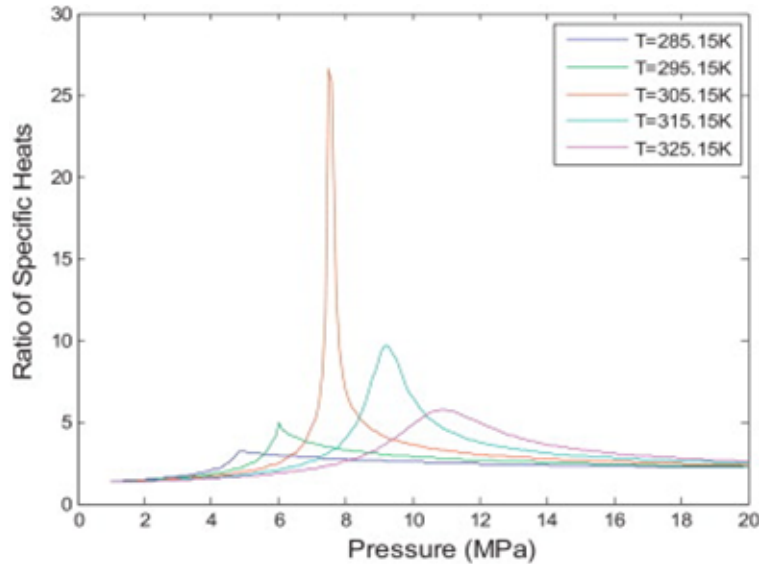


Figure 2-5 Specific heat versus temperature with varying pressures for CO₂ [144]

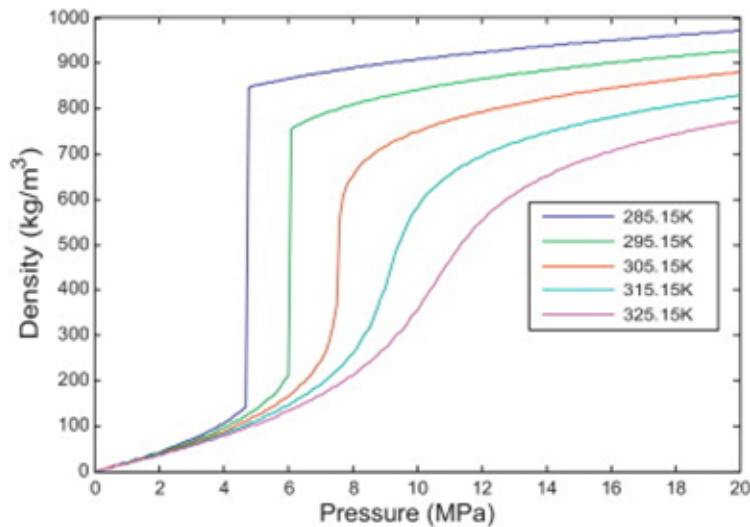


Figure 2-6 Density versus temperature with varying pressures for CO₂ [144]

In Figure 2-7, it is observed that the Brayton cycle locates at the monophasic gas-phase area, but the Rankine cycle varies between vapour and liquid phase. For both cycles, the heat is input and output at constant pressure. Before inputting the heat, the pressure is increased by a pump/compressor. An expander reduces pressure in order to produce work. Moreover, their cycle efficiency can be increased by introducing reheating, intercooling and recompression.

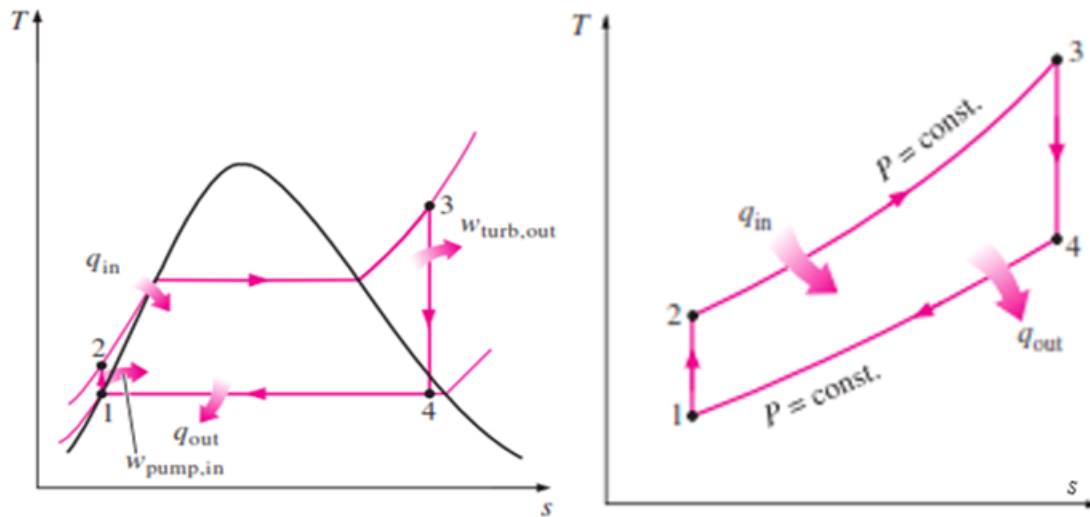


Figure 2-7 T-S diagram of Rankine cycle and Brayton cycle [145]

There are six advantages for sCO₂ power cycle: 1) the supercritical fluid properties near the critical area are able to increase the cycle efficiency. In Figure 2-8, the sCO₂ power cycle has high thermal efficiency at moderate temperatures ranging from 450°C to 700°C by virtue to the less work requirement of CO₂ compression near the critical point; 2) the sCO₂ power cycle is allowed to operate under relatively low pressure by virtue to the critical pressure of CO₂ is one-third of the water, which improves the cycle efficiency; 3) the nearly ambient critical temperature of CO₂ is able to achieve sCO₂ power cycle applications in wide range heat sources and the cycle operates with near-ambient sinks; 4) lower compressor outlet pressure (COP) and high sCO₂ density lead to the compact turbomachinery (Figure 2-9) and the heat exchanger. The size of turbomachinery using sCO₂ as a working medium is 10 times smaller than that using steam as the working medium,

further resulting in a smaller footprint and lower capital cost. In addition, additional auxiliary equipment requires in the CFPP, such as the large heat recovery steam generator (HRSG) and the condenser, which are difficult to minimum their size and take up additional land; 5) in the heat exchanger, that the trajectory of sCO₂ temperature well matches the heat source, which avoids the problem of pinch point and reducing the reversibility; 6) the sCO₂ turbine has a relatively small number of stages by virtue to lower pressure ratio. The critical temperature of sCO₂ can be condensed by air resulting in reducing the impact on the environment compared with water.

Nonetheless, the disadvantages cannot be ignored, such as the material corrosion under high temperature and pressure, 2-3% efficiency improvement per 6.9 MPa from 13.8 to 27.6 MPa of COP [146], and equipment redesign owing to the high-pressure sCO₂.

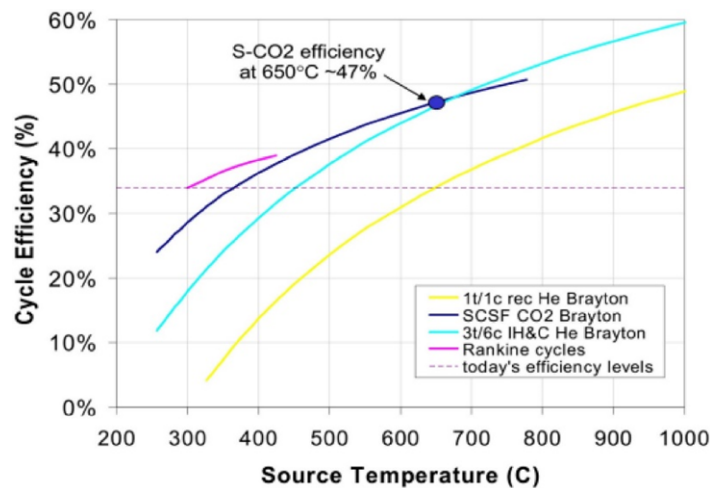


Figure 2-8 Relationship between cycle thermal efficiency with heat source temperature (1t/1c rec He Brayton – one turbine and one compressor; SCFC CO₂ Brayton cycle – recompression sCO₂ Brayton cycle; 3t/6c IH&C He Brayton – 3 turbines and 6 compressors) [147]

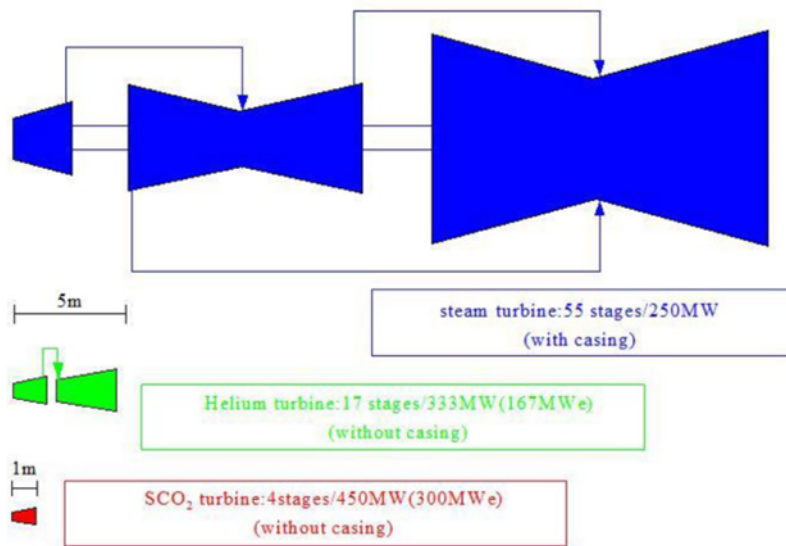


Figure 2-9 Turbine size comparison using helium, steam and sCO₂ as the working medium [148]

The typical sCO₂ power cycle layout consists of principal components [149]– an expander, heat exchangers, a recuperator heat exchanger, a waste heat rejection exchanger and a compressor. The design of these components is particular due to the physical properties, including high pressure, high power density, low viscosity and upheaval on the sCO₂ properties near the critical point. Furthermore, other subsystems such as turbomachinery and BOP are essential for allowing the sCO₂ power cycle as an integral system. For example, the bearing and seal, and the thrust management system for turbomachinery, gear boxes, motor/generators, gas management systems, the waste heat rejection system, piping, skid systems, start-up system components and valves, oil lubrication, house air, instrumentation and control systems, and electrical switch gear for BOP.

The size of the CFPP employing the sCO₂ power cycle is a major challenge. The sCO₂ turbines are theoretically more compact than similar capacity steam turbines, which is non-existent and under designing for large-scale application [150–152]. The 10 MW level load capacity of the sCO₂ recuperator has been built up to now, and the 47 MW recuperator has been designed for a 10 MW sCO₂ demonstration

plant [153]. The lower emission and high efficiency CFPP is typically in the range 400-1000 MW, and the sCO₂ recuperator is expected to have the level in the range 1500-3000 MW.

2.4.2 Layouts of the supercritical CO₂ power cycle

In this section, indirect heated sCO₂ power cycles are described, including recuperated sCO₂ power cycle and recompression sCO₂ power cycle, which are applied in the proposed systems of this thesis. Meanwhile, the direct sCO₂ power cycle, Allam-Fetvedt Cycle (Allam power cycle), is representative in recent years. The Allam cycle is a system that combines the concepts of the open-loop Brayton cycle and the semi-closed oxy-fuel combustion using transcritical CO₂ as the working fluid. It is a process for converting gaseous fuels into thermal energy with the capture of CO₂ and H₂O.

2.4.2.1 Recuperated supercritical CO₂ power cycle

The layout of a simple recuperated sCO₂ power cycle (Figure 2-10) includes a turbine, a compressor, a recuperator, a primary heat exchanger and a primary cooler. The recuperator is used to transfer the heat from the outlet of the turbine to the stream before entering the primary heat exchanger. After the hot side of the recuperator, the CO₂ is cooled and pressurized to its nearly above critical point in the primary cooler and the compressor, separately, which is for preheating in the cold side of the recuperator. The temperature imbalance at the hot end of the recuperator further results in efficiency reduction, owing to the higher heat capacity near the critical point. Modifying to recompression power cycle is one way to compensate, enhancing the heat transfer and improving the cycle efficiency. Another compensation way is by heating a partial high-pressure sCO₂ stream through waste heat from a heat source (such as the low-pressure sCO₂ stream from the hot-end high-temperature recuperator (HTR), in Figure 2-11), which was investigated in Chapter 4.

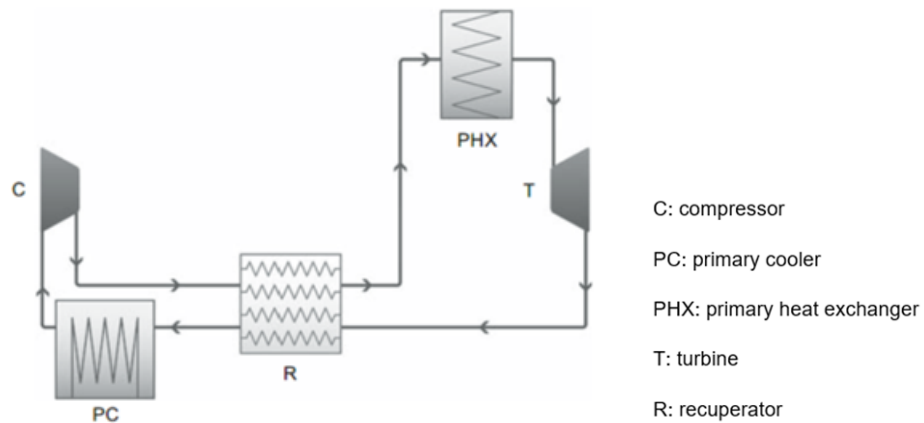


Figure 2-10 Layout of the simple recuperated sCO₂ cycle [39]

2.4.2.2 Recompression supercritical CO₂ power cycle

Based on the simple recuperated sCO₂ power cycle, recompression sCO₂ power cycle (Figure 2-11) employs two recuperators, low-temperature recuperator (LTR) and HTR. A portion of the sCO₂ stream is separated at the end of the hot-side LTR and then mixed with the sCO₂ stream at the end of the hot-side LTR through the recompressor. Another portion of sCO₂ is the beginning of the hot-side LTR via the primary cooler and the main compressor. Then the mixed stream passes the HTR and joins the primary heat exchanger gaining heat for further driving the turbine. The layout of the recompression sCO₂ power cycle aimed to balance the heat duty passing the LTR, ensuring a minimum temperature approach between both ends of the LTR and the cold end of the HTR [39].

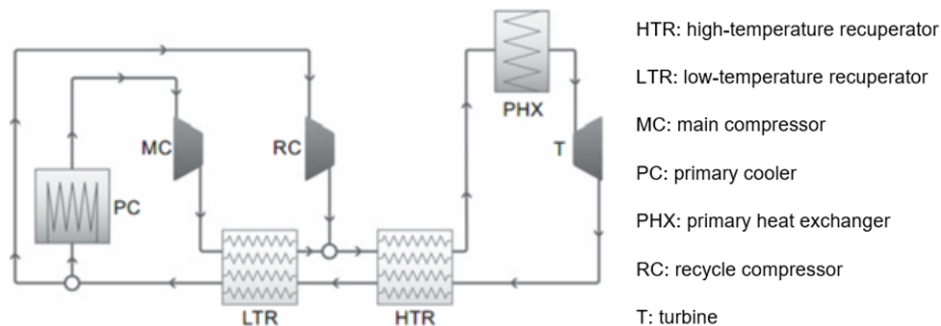


Figure 2-11 Layout of the recompression sCO₂ cycle [39].

The recompression sCO₂ power cycle applying on the coal-fuelled power plant is complicated due to the small temperature window used for heat absorption, comparing with its application in concentrated solar power plants and nuclear power plants. In addition, the significant remaining heat in the flue gas after the primary heat exchanger should be considered for effective use, such as the additional advanced air preheaters studied in Chapter 5, or sCO₂ bottoming cycles, cogeneration and combined heat and power, steam-Rankine bottoming cycles.

2.4.3 Integration with conventional and nonconventional coal-fired power plants

Careful consideration is required for integrating the sCO₂ power cycle with the CFPP. The designs of the sCO₂ primary heater has significant differences comparing with the incumbent primary heater in the steam Rankine cycle. First, the required mass flow rates of sCO₂ are around 10 times greater than that of steam based on the same level of heat input which is the attribute to the lower sCO₂ turbine pressure ratio and around 3 times lower mass-basis specific heat of CO₂ over the heater/boiler's temperature and pressure ranges. At last, the heater tube sizing has been significantly impacted, which results in a further difference – the reasonable pressure drop. In the sCO₂ power cycle, the energy requirement to compress sCO₂ is significantly higher than that to compress steam, and minimizing pressure drop of piping is able to improve the efficiency of the cycle and the plant. The last difference is that the sCO₂ power cycle has a different temperature heat input window compared with the steam cycle.

Regarding the integration with the conventional CFPP, sCO₂ primary heater has many differences compared with the steam primary heater, but some of the combustion processes generally remain the same, such as the processes of coal handling, ash handling, and low NO_x burner. In 2014, a sCO₂ primary heater was investigated by EPRI and Babcock & Wilcox for a CFPP [154]. It includes a sCO₂ recompression power cycle and an inverted tower boiler for adapting the sCO₂ power cycle in the higher temperature operation, shown in Figure 2-12. The study

indicated that lower pressure drop achieved large tube diameters when using nickel material operating over 620°C, resulting in a severe impact on the boiler cost. In addition to the above, the flue gas temperature is about 566°C, due to the smaller temperature window for heat absorption into the sCO₂ power cycle, which significantly exceeds the capability of the incumbent flue gas air preheater with flue gas temperature of about 370°C. In addition to the above, due to the smaller temperature window of heat absorption in the sCO₂ power cycle with the flue gas temperature of 566°C, the incumbent flue gas air preheaters do not have enough capabilities to transferring heat with the temperature of about 370°C. Therefore, a cascade sCO₂ bottoming cycle is added for reasonably reducing the flue gas temperature. Furthermore, the refractory lining of heater walls is employed after the membrane wall because the membrane wall cannot be efficiently cooled owing to the higher inlet temperature into the sCO₂ primary heater.

A conceptual study from Moullec [155] developed a simple sCO₂ primary heater for a CFPP employed sCO₂ recompression power cycle with double reheat and maximum operating temperature of 620°C, further integrated with the MEA-based CCS process. The study found the overall pressure drop through the main heat, first reheat and second reheat is 18.3 bar. The pressure reduction of the membrane wall occupies 59% points, the main heat economizer occupies 14%, and the remains occur in the 6 sets of superheat tubes. If the mentioned pressure reductions are disregarded, the net plant thermal efficiency in lower heating value could be increased by 1.6%. Also, the issues of high flue gas temperature to 540°C are mentioned, and thus a high-temperature air preheater and a parallel economizer are added for sufficiently cooling the flue gas to 110°C for clean-up. Although his additional study investigated alternative configurations for tackling the high flue gas temperature [156], the original study [155] is ultimately recommended. On the whole, both studies show the 2.4-4.9% points increase in the net plant efficiency in LHV comparing with the steam Rankine cycle.

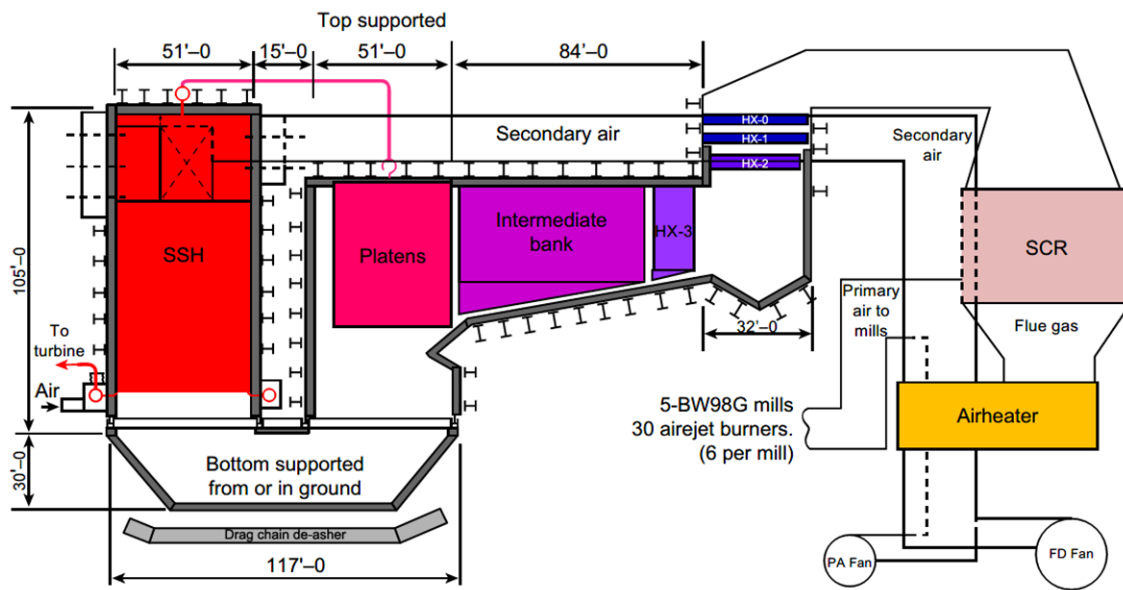


Figure 2-12 The inverted tower boiler for sCO₂ power cycle [154]

Regarding the integration with the nonconventional CFPP, some of the steam boilers are able to accommodate well the sCO₂ power cycle, such as fixed bed, BFB and CFB, lying on the flow rates of air past the bed. The CFB and the BFB have been commercially applied by fulling coal and biomass, respectively [157]. A significant advantage of CFB combustion is the sulphur removal by adding limestone and coal to the combustor. The limestone finally generates the calcium sulphates and then removing with the coal ash, which does not need the sulphur removal process the downstream resulting in the benefit regarding the cost and efficiency. The required sulphur removal occurs in the combustor operating between 800°C and 900°C, which also prevents the formation of NO_x with its associated equipment. The higher flue gas temperature is able to be recycled to the combustor due to the lower temperature window of sCO₂. The coal boiler is able to operate in air- and oxy-combustion without significant modifications.

NETL has studied a sCO₂ recompression power cycle integrating with an oxy-coal combustion CFB boiler with CCS [158]. The system uses the remaining heat from the exhaust gas to preheat the recycled flue gas and partial stream of the sCO₂ power cycle. In addition, the study examines several sCO₂ power cycles. At 620°C

of TIT, comparing with the Rankine cycle, the sCO₂ power cycle with reheat and intercooling shows a 2%-point efficiency improvement. The additional TIT of 760°C is investigated, achieving a net plant thermal efficiency of 39.3%_{HHV}. A Zero Emission system was designed by integrating the coal-fired oxy-combustion pressurized fluidized bed combustion (PFBC) boiler with the sCO₂ recompression power cycle, and a flue gas turbine is introduced for the extra power production, shown in Figure 2-13 [159,160]. The PFBC is a BFB operating at 0.83 MPa and 871°C with the in-bed heat exchange tubes for heat transfer with sCO₂ in the PFBC [161]. The results indicated 2.4-3.7% points increase in net plant efficiency in HHV comparing with the steam Rankine cycle. Recently, a pilot-scale PFBC exchanging heat with sCO₂ was constructed and tested in collaboration between the Gas Technology Institute and the CANMET [162]. In addition, a potential cost-effective coal-fired oxy-combustor in a sCO₂ environment at 10 MPa has been integrated with a sCO₂ power cycle [163]. Nonetheless, significant considerations should be a notice on the designs for such as the high temperature and pressure of combustor in sCO₂ environment, ash particulate removal in the cyclone, and wear protection of recuperators.

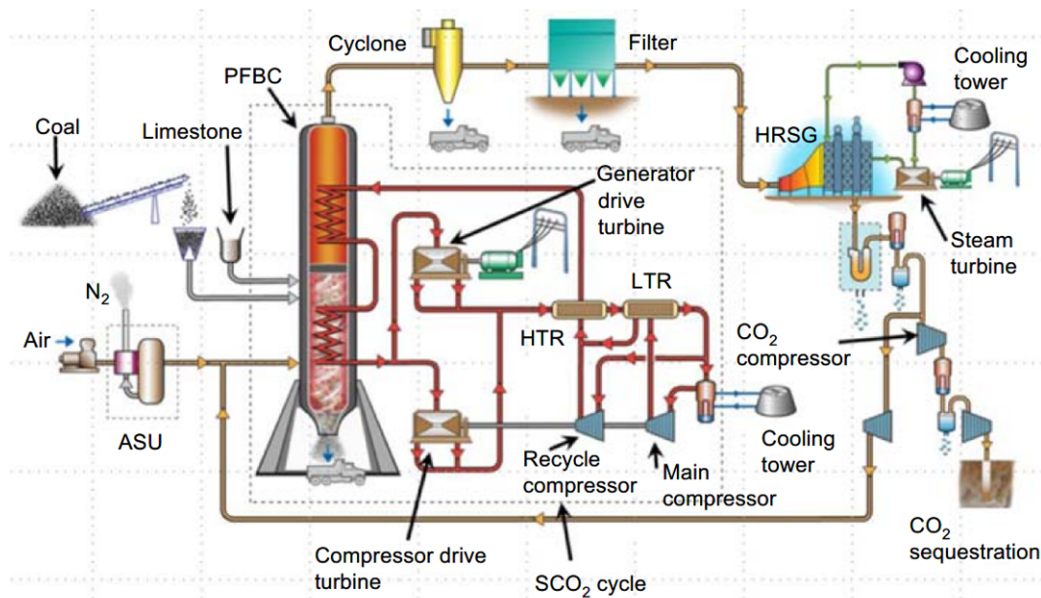


Figure 2-13 The Zero Emissions system [159]

To sum up, the sCO₂ recompression power cycle used to be integrated with the coal-fired primary heater. Further research should be conducted for the most applicable sCO₂ power cycle accommodating coal combustion. The major challenges include the utilisation of high flue gas temperature, minimization of pressure drop in PHX and improvement on the power density of the sCO₂ power cycle for reducing its overall size.

2.4.4 The economics of supercritical CO₂ power plants

2.4.4.1 Potential markets in fossil fuel power plants

The first implementation of the sCO₂ power cycle in a natural gas power plant has 150 MW_{el} capacity with around 50% net cycle efficiency at TIT 700-750°C [164]. The capital cost is expected to be around \$1000/kW_{el} considering the recompression sCO₂ power cycle with its variations of reheating or intercooling, and additional effective technologies such as inlet air preheating. In a fast-growing market, due to economies of scale [165], investing in large plants is reasonable can achieve the lower capital cost [166]. However, in a slow-growing market (the United States), the smaller plant is more popular [166]. Furthermore, the plants are able to operate at unprecedented efficiencies by firing gaseous or liquid fossil fuel.

Furthermore, direct and indirect oxy-fuel combustion power plants are considered to implement the sCO₂ power cycle. Direct oxy-fuel combustion power plants are revolutionary by virtue to the concepts of CCS. The plant is called the Allam cycle, and its patent belongs to Net Power in North Carolina [167]. The ASU separated O₂ from air consumes 6-7% of the gross power. Afterwards, the fuel combusts with O₂ and is furthering compressed to around 30 MPa. The sCO₂ then recirculates for controlling combustion chamber temperature. The main combustion products are CO₂ and H₂O without NO_x is generated. In the process, the mixture in the sCO₂ power plant was heated to 1100-1200°C by the injected high-pressure combustion products. The cost of the plant could be significantly reduced, and the efficiency is able to rise over 60%. In the turbine, the CO₂/H₂O mixture is expanded to 8-9 MPa

at a pressure ratio from 3 to 3.5. After condensing the water from the mixture, the CO₂ is able to deliver for EOR or CCS without an additional compression process. For this type of power plant, the high efficiency, needless high-temperature HX, and zero emissions are major economic benefits when burning fossil fuels. However, attention should be paid to the risks of the high-pressure and high-temperature combustor and the turbine. The power density of the turbine is approximately equivalent to that of the rocket engine. If the system aims to achieve economic feasibility, operation of the turbine for many long times at higher pressure is necessary. Currently, NET Power is actively evaluating the location of the first commercial-scale 300 MW facility. Indirect oxy-fuel combustion plants are similar to the direct oxy-fuel combustion plant referencing the concept of CCS. The indirect oxy-fuel combustion has a quantity of variants. The high-pressure variant uses a similar combustion process with direct oxy-fuel combustion. However, the combustion temperature and the pressure are estimated to be 800-1000°C and 10-12 MPa, respectively. Then, the high-temperature and high-pressure exhaust gas exchanges heat with sCO₂ in the closed-loop system. The sCO₂ TIT is heated to 700-750°C with an efficiency of 50%. Due to the high pressure and density of the combustion gas, the PHX is smaller than the typical. Afterwards, the combustion gases are expanded in a turbine probably with the outlet pressure of 8-9 MPa, and then are reinjected into the high-pressure combustion chamber for controlling its inner temperature. The waste heat in the gases can be used to lower the energy penalty from the ASU. Finally, after the gases are condensed by removing the water, the CO₂ at a pressure of 8-9 MPa is delivered for EOR and CCS, similarly, without an extra energy-intensive compression process. The net efficiency of the indirect sCO₂ power cycle is expected to be 42-43%. From the kinds of literature, Natural Resources Canada [168] performed the original work for this type of plant, and the recent work has been done by industry and institutes [169,170].

A technical report from Sandia National Laboratories [171] estimated LCOE of the sCO₂ power cycle system are between 44.8 and 56.1 \$/MWh in the plant size

range from 100 to 300 MW_{el}. Meanwhile, the report pointed out that there are many uncertainties to commercialization, such as a lower cost needs to be demonstrated in the long-term during operation, and the ability to quickly integrating with intermittent resources should possess. In addition, a technical report from Echogen Power Systems (DE), Inc [172] achieves 216.9 \$/MWh of LCOE in the case of the CFPP integrating with the sCO₂ power cycle and CCS. If that case does not use CCS, the net efficiency and LCOE are 40.3%_{HHV} and 155.4 \$/MWh, respectively. Park et al. [173] performed the net electricity efficiency of a 625 MW coal-fired power plant with the sCO₂ Brayton cycle was 6.2–7.4% higher than that of the steam Rankine cycle with a 7.8–13.6% reduction of the LCOE.

2.4.4.2 Key performance indicators

This section introduces reference cases with crucial performance indicators evaluating economic feasibility, including LCOE, internal rate of return (IRR) and net present value (NPV). However, in this thesis, only LCOE is primarily introduced in Section 3 with application in the studied cases of Sections 4, 5 and 6.

Taking a combined cycle plant as an example, a General Electric LM-2500PE gas turbine is paired with a waste heat recovery power system using the simple recuperated sCO₂ Brayton cycle. The nameplate capacity of the plant is 32,017 kW_{el}, with the annual utilization rate of 85% and the lifetime of 20 years. The 'Project cost' is estimated to \$30,797,000 with the \$10,778,950 tax shields (35% tax rate) during its lifetime. The major operating cost is the fuel (natural gas) cost of \$0.03662/kWh_{el}. Other operating and maintenance cost is estimated to \$0.008/kWh_{el}. The final LCOE is calculated to \$0.0409/kWh_{el}. LCOE is an efficient way to competing among technologies, using combined cycle power plant with a simple recuperated Bryton cycle is economically reasonable and competitive, as long as the LCOE of the comparative projects is lower than \$0.0409/kWh_{el}. This example assumes several parameters, such as inflation rate, depreciation year, business tax rate, CO₂ tax and cost of fuel. In addition, the local policy is able to impact the assumption by furthering influencing the final economic results. So, the

same assumptions have to be used for effectively comparing among projects. The IRR is a key performance indicator for investigators determining the economic viability of the project. If the interest rates are below the IRR, a positive rate of return can be achieved. IRR is the discount rate that the present value of the future cash flows equals zero, or that a project reaches the break-even point. Taking the aforementioned combined cycle power plant with a simple recuperated Bryton cycle, the sales price of electricity is assumed to \$0.06/kWh with final annual revenue of \$14,303,900. Annual operation costs and annual depreciation expenses are \$10,636,258 and \$3,079,700, respectively, calculated from the LCOE. Tax expenditures account for 35% of the net annual profit. During the first decade, depreciated annual cash flow was \$382,171.77. In addition, the free cash flow is \$3,461,871.77 for the first 10 years by calculating the tax expenditures. Once it is depreciated, taxable profits rise to \$3,667,656.57 with tax expenditures rise to \$1,283,679.80, and annual free cash flow decreases to \$2,383,976.77. The final IRR is calculated to 7.924% for the combined cycle power plant, which means the project is economically viable when interest rates are below 7.924%. The next step uses NPV for estimating value. The formulas of NPV and IRR are similar, but with the difference in the formula NPV, the interest rate of the debt-funded for the project would be used to solve NPV. The NPV of the combined cycle is calculated to \$7,235,839 by assuming the average capital cost of the project to be 5.0%, which means that, apart from the initial investment is gets back, the investors will earn \$7.2 million during the 20-year lifetime of the project.

2.4.4.3 Cost of components

For providing an estimation of the project cost in the economic estimation for such power system in Section 2.4.4.2, there is a 10 MW_{el} sCO₂ power system was estimates by Wright et al. for the cost of components. HXs account for most of the cost of sCO₂ power systems. So size optimization of HXs reduces the cost of the sCO₂ power plant. The costs of turbomachinery, auxiliary equipment and BOP were calculated together in one group.

In terms of HXs (including the PHX, the preheater, the HTR, the LTR and the CO₂ condenser), their costs are in direct ratio with $U \times A$ with the equation of $U \times A = Q / \text{LMDT}$ (U: universal heat transfer coefficient; A: area; Q: transferred heat in the heat exchanger; LMDT: log mean differential temperature). The cost of heat exchangers increases with the increase of heat duty and decrease of LMDT.

The costs of turbomachinery with BOP components, such as the turbine, the compressor, the bearing and seal, gearbox system, generator, motor, piping, control system and water cooling system. These cost estimations have not included the developer's non-recoverable engineering (NRE) costs, but have included the NRE costs of the original equipment manufacturers (OEM). Therefore, it is better to predict the first of a kind (FOAK) sCO₂ system for the cost estimations, rather than the nth of a kind (NOAK) plant, due to the cost of the FOAK is able to be significantly higher than that of the NOAK.

2.5 Research gaps

From the above introductions, the combination of CLC and the sCO₂ recompression power cycle was placed great expectations on improving the system techno-economic performance. Also, different types of OCs were investigated as they are an essential uncertainty in CLC technology. Through applying the assessment methods in Chapter 3, this work could give the reference for future research regarding CLC and sCO₂ power cycle from a techno-economic perspective.

3 Methods

3.1 Software selection

AspenTech software has been used to design and optimise technical processes to maximize the return during the plant's life cycle. The software products for the manufacturing and supply chain allow companies to increase profits in factories and supply chains. Combining these two product lines creates the Enterprise Operations Management (EOM) solution, an integrated enterprise system that enables process industry manufacturers, to significantly improve operational performance.

In the late 1970s, a project named Advanced Process Engineering Systems (ASPEN) involved MIT and the USDOE and developed the third generation of process simulation software Aspen Plus™. It is used in large-scale multi-purpose process simulation systems for production plant design, steady-state simulation and optimization, and is one of the core software products in AspenTech. The project was completed in late 1981. The following year, AspenTech was established for the commercialization of the software and named Aspen Plus™. Over the past decades, more than 10 versions have been updated, and it is recognized as standard large-scale process simulation software worldwide with millions of applications. Many of the world's leading process manufacturers in the chemical, petrochemical, and refining industries and well-known engineering companies use Aspen Plus™. Aspen Plus™ offers proven and substantial economic benefits throughout the process life cycle from research and development. It brings steady-state model functionality to the engineering desktop, delivering an unparalleled combination of model functionality and ease of use. Aspen Plus™ enables the design, simulation, troubleshooting, and management of productive production facilities.

Aspen Plus™ is recognized as having the most complete physical property system for industry, allowing for more accurate and reliable simulation results. Many

companies standardise their physical properties by using Aspen Plus™'s physical properties system in conjunction with their engineering computing software. In Table A-1, Aspen Plus™ physical property databanks including pure components, solids, aqueous, binary, combustion and inorganic. Aspen Plus™ is the only software approved to interface with the DECHEMA (German Society for Chemical Apparatus) database. The database collects more than 250,000 sets of the world's most complete data on gas-liquid equilibria and liquid-liquid equilibria. Users can also connect their physical data to the Aspen Plus™ system.

Two algorithms, the sequential module (SM) and the equation oriented (EO), are uniquely included in a simulation tool. The sequential algorithm provides the initial value of the process convergence calculation, and the simultaneous equation algorithm dramatically improves the convergence speed of large-scale process calculation. At the same time, it makes the previously complex process calculation possible, which saves time for engineers.

In addition to component, property and equation of state, it also includes the following unit operation model for gas/liquid system and solid system, shown in Table A-2.

Further, Aspen Plus™ offers a powerful set of modelling tools to maximize the benefits of process modelling. Convergence Analysis automatically analyses and recommends optimized tear flow, process convergence methods and computational sequences, even for huge processes with multiple streams and information cycles. Calculator Models contains ad-hoc computing, and embedded FORTRAN and Microsoft Excel model interfaces. Sensitivity Analysis easily tabulated and graphed process parameters varying with equipment specifications and operating conditions. Case Study performs multiple simulations, comparisons, and analyses with different inputs. Design Specification automatically calculates operating conditions or device parameters to meet specified performance objectives. Data fitting is that using the process model fits with real device data to ensure accurate and effective real device models. Optimization determines the

operating conditions of the unit to maximize any specified objectives such as yield, energy consumption, logistics purity, and process economy.

3.2 Techno-economic assessment

The proposed cases of oxy-fuel combustion and CLC in this thesis have been considered and their thermodynamic and economic performance was assessed. The method for the techno-economic assessment (TEA) employed in this work was based on the past work of Hanak et al. [174] and further considered refinements based on the work of Ciferno et al. [175] and Michalski et al. [176]. These were presented in detail in Section 3.2.1 and Section 3.2.2.

3.2.1 Thermodynamic assessment

To evaluate the thermodynamic performance, the process models for the considered cases have been developed in Aspen Plus™. One of the key performance indicators is the net electricity efficiency of the entire power plant (η_{net}), which is the ratio of the net power output of the entire power plant and heat input from fuel combustion, as shown in Eq. (1). The net power output of the power plant is the difference between the gross power output (P_G) generated by the generator driven by a turbine, and the total power consumption of the auxiliary equipment ($\sum P_{AUX}$) in the power plant. In oxy-fuel combustion cases, $\sum P_{AUX}$ mainly comprises power consumption of the induced draft fan, the forced fan, the ASU and the CPU. Conversely, the CLC does not include the power consumption from the ASU. The energy input from fuel combustion was calculated by the product of the mass flow rate of the fuel ($\dot{m}_{fuel}$) and higher heating values (HHV) of the fuel that was estimated by the Dulong formula [177]. Specific CO₂ emission (e_{CO_2} , Eq. (2)) is the main environmental performance indicator in the context of global efforts to reduce emissions, which is known as the ratio of CO₂ emission rate ($\dot{m}_{CO_2}$) and the net output power of the power plant ($\dot{W}_{net}$).

$$\eta_{net} = \frac{P_G - \sum P_{AUX}}{\dot{m}_{fuel} \cdot HHV} [\%] \quad (1)$$

$$e_{CO_2} = \frac{\dot{m}_{CO_2}}{\dot{W}_{net}} [g_{CO_2}/kW_{el}h] \quad (2)$$

The main input streams include mass flow rate of actual air input into the air reactor and mass flow rate of actual oxygen carrier circulation [115,178] and mass flow rate of gasification agent from recycled flue gas, both estimated in the Aspen PlusTM model. Besides, both the mass flow rate of OC makeup and solid fuel are estimated by the reference report [44]. A key parameter, oxygen transport capacity (R_O) (Eq. (3)), was introduced representing the ability to carry the O_2 of an OC. The m_{ox} and m_{red} represent the sample mass in fully oxidized and fully reduced form. Char conversion efficiency in the FR ($X_{char,FR}$, Eq. (4)) is explained as unconverted C_{fix} fed into the AR to C_{fix} in the solid fuel.

$$R_O = \frac{m_{ox} - m_{red}}{m_{ox}} \quad (3)$$

$$X_{char,FR} = \frac{\text{Unconverted fixed carbon fed into AR}}{\text{Fixed carbon in the solid fuel}} [\%] \quad (4)$$

3.2.2 Economic assessment

The economic performance of the proposed cases is compared with the reference cases, respectively, for oxy-fuel CFPP [98] and chemical looping combustion CFPP [178]. The LCOE is widely used to estimate the cost of electricity shown as Eq. (5) [98], defined as the ratio of the total cost and the total power output of the power plant during its lifetime. It includes total capital requirement (TCR), fixed charge factor (FCF), fixed operating and maintenance cost (FOM), capacity factor (CF), variable operating and maintenance cost (VOM) and specific fuel cost (SFC), which considers the lifetime of the power plant and project interest rate. It is worth noting that the cost associated with CO_2 emissions is accounted for in the VOM, calculated as the product of carbon tax and annual CO_2 emissions rate.

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

The cost of CO₂ avoided (AC, Eq. (6)) quantifies the average cost of avoiding a unit of atmospheric CO₂ emissions when delivering a unit of electricity [179], which compares a plant with CCS to a reference plant without CCS. The cost of CO₂ captured (CC, Eq. (7)) is an economic parameter to assess the economic viability of CO₂ capture systems compared to the CO₂ price in the market. As the above two parameters, there are two details worth to be noticed: 1). The reference power plant for these two calculations uses Case 11 (a supercritical pulverized coal steam cycle power plant) in a report [175] from USDOE/NETL; 2). The LCOE_{CCS} in the AC includes the total cost of CCS. Rather than, in the CC, the LCOE_{CC} excludes the CO₂ transport and storage cost. It is because the CC is a quantitative measure compared to the commercial market price of CO₂, such as the food industry and the petroleum industry.

$$\text{Cost of } CO_2 \text{ avoided} = \frac{LCOE_{CCS} - LCOE_{ref}}{e_{CO_2,ref} - e_{CO_2,CCS}} \text{ [€/tCO}_2\text{]} \quad (6)$$

$$\text{Cost of } CO_2 \text{ captured} = \frac{LCOE_{CC} - LCOE_{ref}}{e_{CO_2,capture}} \text{ [€/tCO}_2\text{]} \quad (7)$$

The capital cost of the power plant is estimated using the economic data from the reference power plant (Table 3-1 and Table 3-2) via the exponential method function [180]. The main difference between the considered cases is the type of power cycle. Therefore, the capital costs of the power cycle and the cooling tower system needs to be recalculated in the cases that consider the sCO₂ cycle. This is achieved using Eq. (8) that consider the bare erected costs (BEC, M€), engineering, construction management, home office and fees (*Eng'g CM H.O. & Fee*, M€), process contingency (*Process Cont.*, M€) and project contingency (*Project Cont.*, M€) [175].

$$TCR = BEC + Eng'g \text{ CM H.O. \& Fee} + Process \text{ Cont.} + Project \text{ Cont.} \quad (8)$$

As there is no reliable data source for the cost of the sCO₂ cycle, Eq. (9) is used to estimate its BEC. This depends on the BECs of the supercritical CO₂ compressor (C_C), the CO₂ expander (C_E), four heat exchangers (ΣC_{HE}), the electric generator (C_{EG}) and the cooling tower (C_{CT}), and the piping and integration cost indicator ($i_{P\&C}$).

$$C_{cycle,CO_2} = (1 + i_{P\&C}) \cdot (C_C + C_E + \Sigma C_{HE} + C_{EG} + C_{CT}) \quad (9)$$

The supercritical CO₂ compressor and the expander are the two main unit operations of such the cycle. Their BECs are estimated using the methodology developed by Benjelloun et al. [181] shown in Eq. (10). The BEC of the supercritical CO₂ compressor is determined by the compressor inlet pressure (p_{in}), compressor isentropic efficiency ($\eta_{i,Com}$), equivalent air mass flow rate ($\dot{m}_{AE,Com}$), and equivalent air outlet pressure ($p_{AE,out}$).

$$C_C = \dot{m}_{AE,Com} \cdot \frac{47.1}{1 - \eta_{i,Com}} \cdot \frac{p_{AE,out}}{p_{in}} \cdot \ln\left(\frac{p_{AE,out}}{p_{in}}\right) \quad (10)$$

In terms of the BEC of the CO₂ turbine, it depends on the expander isentropic efficiency ($\eta_{i,Exp}$), equivalent air mass flow rate ($\dot{m}_{AE,Exp}$), equivalent air outlet pressure ($p_{AE,out}$) and expander inlet pressure (p_{in}) (Eq. (11)). The constant 392.2 was derived based on the results from Criado, Arias and Abanades [182], and Gabbrielli and Singh [183].

$$C_E = \dot{m}_{AE,Exp} \cdot \frac{392.2}{1 - \eta_{i,Exp}} \cdot \frac{p_{in}}{p_{AE,out}} \cdot \ln\left(\frac{p_{in}}{p_{AE,out}}\right) \cdot [1 + \exp(0.036 \cdot T_{in} - 65.66)] \quad (11)$$

To calculate $\dot{m}_{AE,Com/Exp}$, the volume flow rate of air and CO₂ at the inlet of the compressor and the expander were assumed to be equal, and the mass flow rate is proportional to the density, as shown in Eq. (12). Assuming that the outlet

volumetric flow rate of CO₂ and air are also the same, the outlet air density ($\rho_{out,Air}$), in the case of equivalent outlet air pressure of the compressor and the expander, are derived (Eq. (13)).

$$\dot{m}_{AE,Com/Exp} = \dot{m}_{CO_2,Com/Exp} \cdot \frac{\rho_{in,Air}}{\rho_{in,CO_2}} \quad (12)$$

$$\rho_{out,Air} = \frac{\dot{m}_{AE,Com/Exp}}{\dot{m}_{CO_2,Com/Exp}} \cdot \rho_{out,CO_2} \quad (13)$$

To obtain the air density, the corresponding equivalent outlet pressure was determined using an iterative process. It needs to be highlighted that all calculations performed in this work with Eq. (12) and (13) used the real gas database (CoolProp) [184].

The BECs of heat exchangers (C_{HE}) including the superheater, the recuperator, the economiser and the cooler were estimated using Eq. (14) which depends on the heat exchange surface (A_{HE}) and highest pressure of the heat exchanger (p_{HE}). The BEC of the electric generator (C_{EG}) used the gross power output of the generator (P_G) as the scaling factor in Eq. (15) [185].

$$C_{HE} = 2546.9 \cdot A_{HE}^{0.67} \cdot p_{HE}^{0.28} [\text{€}] \quad (14)$$

$$C_{EG} = 84.5 \cdot P_G^{0.95} [\text{€}] \quad (15)$$

The BEC of the cooling tower system (C_{CT}) was determined considering its unit cost and heat duty ($\dot{Q}_{CT}$) as presented in Eq. (16). The unit cost of the cooling tower system was set to 32.3 €/kW [175].

$$C_{CT} = 32.3 \dot{Q}_{CT} [\text{€}] \quad (16)$$

Furthermore, the process contingency is assumed to be zero. Eng'g CM H.O. & Fee and Project Contingency (M€) for the considered cases were estimated using

proportions based on their corresponding BECs shown in Table 3-1 and Table 3-2. Also, since feedwater and miscellaneous BOP systems are not applicable in proposed cases, these are not counted for in the TCR. Such for the oxy-fuel combustion cases, the annual cost rate of variable operations and maintenance (O&M) is assumed to be 2.0% of TCR, and the annual cost rate of fixed O&M is 1.0% of TCR. The fixed charge factor, which is defined as a ratio used to determine the present value of an annuity, is about 10% without considering salvage value, tax and depreciation [51]. The base year for economic calculation was 2018 in Chapter 4 and 2019 in Chapter 5. It is also assumed that the variable costs are constant over time in real terms.

Table 3-1 Economic model assumptions for oxy-fuel combustion with sCO₂ power cycle in Chapter 4

Parameter	Value
<i>Reference oxy-fuel combustion power plant</i>	
Reference total plant cost (M€) [186]	1302.2
Reference gross power output (MW) [186]	785.9
Reference total steam cycle cost (M€) [186]	120.3
Bare erected cost (M€) [186]	98.6
Engineering, Construction Management, Home Office & Fees [186]	9.1 (9.23% of BEC)
Project contingency (M€) [186]	12.5 (12.70% of BEC)
Reference total feedwater & misc. BOP systems cost (M€) [186]	78.1
Bare erected cost (M€) [186]	61.6
Engineering, Construction Management, Home Office & Fees (M€) [186]	5.6 (9.33% of BEC)
Project contingency (M€) [186]	10.9 (14.84% of BEC)
Reference cooling tower system cost (M€) [186]	35.9
Scaling factor [186]	0.67
<i>Other economic parameters</i>	
Variable O&M yearly cost rate (%) [187,188]	2.0
Fixed O&M yearly cost rate (%) [187,188]	1.0
Carbon tax (€/t) [187,188]	0.0
Specific CO ₂ transport and storage cost (M€/a) [98]	7.0
Fuel price (€/GJ)	
Bituminous (South Africa) [189]	2.8
Wood Pellets [190]	4.5

Lifetime (years) [187,188]	25
Real discount rate (%) [187,188]	8.78
Annuity factor (%) [187,188]	10.0
Year time (hours) [187,188]	8760
Capacity factor (%) [187,188]	80

It is worth noting that the thermodynamic assessment is a necessary process to achieve economic performance, due to there are plenty of data (shown in Table B-1 and Table B-2) need from the thermodynamic assessment to feed into the economic assessment.

Table 3-2 Economic model assumptions chemical looping combustion with sCO₂ power cycle in Chapter 5

Parameter	Value
Reference CDCLC power plant	
Total capital requirement (M€) [178]	1456.3
Total steam cycle cost (M€) [178]	143.0
Feedwater & misc. BOP systems cost (M€) [178]	88.6
Cooling tower system cost (M€) [178]	40.345
HRSG (M€) [178]	59.7
Scaling factor [191]	0.67
Other economic parameters	
sCO ₂ power cycle cost	
Eng'g CM H.O. & Fees rate (%) [172]	8.87%
Process Cont. rate (%) [172]	15%
Project Cont. rate (%) [172]	15%
Owner's cost rate (%) [172]	21%
Variable O&M yearly cost rate (%) [178]	2.95%
Carbon tax (€/t)	0.0
Specific CO ₂ transport and storage cost (€/tCO ₂) [98]	7.0
Fuel price (€/GJ)	
Illinois No.6 coal [178]	2.48
Wood Pellets [192]	4.5
Lifetime (years) [178]	25
Real discount rate (%) [178]	11.64%
Annuity factor (%) [178]	12.43%
Capacity factor (%) [178]	85

3.3 Probabilistic assessment

In reality, many problems cannot be clearly expressed by mathematical models due to randomness and complex formulation, which is able to be effectively solved

by a method of Monte Carlo simulation. Physicists at the Los Alamos laboratory first used the Monte Carlo simulation method during the World War II. This method is successful in solving the problem of neutron fission and has been widely used in the field of operational research.

The Monte Carlo simulation is used to simulate the probabilities of different results in a process that cannot be easily predicted because of the intervention of random variables. In Monte Carlo simulation, random values of variables are drawn from a certain probability distribution, so that the random variables have the same distribution characteristics as the actual population. The investigated problems are then analysed using these randomly drawn variables. In the economic evaluation of a project, there are generally several uncertain input variables, and each variable has several possible values for discrete distribution or infinite possible values for a continuous distribution. It is impossible to analyse and calculate all these values. The population parameters can be estimated by taking a sample containing several elements of the population, such as mathematical expected value and variance, and then the population parameters can be estimated or simulated by using the sample parameters. The Monte Carlo simulation method is one such simulation method.

The probability distribution of the random variables (input variables) is obtained by prediction and estimate. A random number generator can be used to generate a series of random numbers with the same probability and statistical properties as the predictor variables, and then map to the distribution function of each random variable through a certain correspondence relationship to generate random values with the same probability distribution. Regardless of the time sequence of their generation, the chances of each value being generated are equal. These random values were used as input variables to calculate output variables, which corresponds to a situation that may occur. The calculation accuracy of the Monte Carlo simulation depends on the number of simulations. When the accuracy is increased by one order of magnitude, the number of calculations must be increased by two orders of magnitude. Therefore, the more repeated calculations,

the distribution of the output will be close to the true distribution and the simulation value will be closed to the true value.

The economic performance of the oxy-fuel combustion and CLC power plant with sCO₂ power cycle are dependent on the assumptions in the economic models. The deterministic model is able to offer dependable predictions for the studied system under any economic assumptions, but ignores the stochasticity of the input variables within reasonable ranges in the actual engineering system and economic environment, such as the variables shown in Table 3-3 for this study. Therefore, a deterministic economic evaluation introduced in Section 3.2.2 cannot comprehensively represent the economic results of an actual system affected by the stochasticity of the economic parameters. To fill the gap, the stochastic approach is introduced into the economic analysis method, thus considering the uncertainty of input variables.

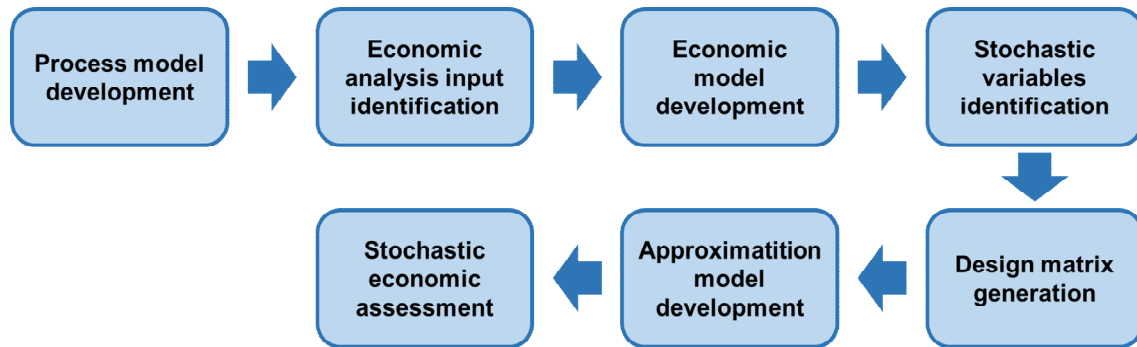


Figure 3-1 Stochastic economic assessment framework [193]

In Figure 3-1, the stochastic economic assessment framework used in this study comprises seven definitive stages referencing the earlier study by Hanak et al. [193–195]. At the first stage, the process models of each technology (in Section 4 and 5) have been developed in Aspen Plus™ with their validation, and the TIT and COP are assumed to be 593°C and 240 bar in all considered cases. At the second stage, the economic calculation method is decided, and the inputs are identified including the stochastic variables. Then, following the economic calculation method, the economic model is established and developed in Microsoft Excel. Next, from the economic model, representative stochastic variables are selected. Then, the

matrix is designed based on Monte Carlo simulations, presented by the stochastic variables and economic indicators with many iterations. Each dataset represents a unique scenario. In this study, there are ten thousand input datasets of input variables are randomly generated following their assumed distributions for the estimation of key economic performance indicators. Afterwards, the statistical results regarding the key economic performance indicators of LCOE, AC and CC are achieved, such as the probability density curves and the key probability indicators.

In Table 3-3, the selection of the stochastic variables has been conducted based on the key assumptions in the economic calculation models, which has the potentials to impact the final economic performance. They are assumed to be uniformly distributed, including the carbon tax, CO₂ transport, storage, and monitoring (TS&M), discount rate, OC price (only considered in CLC case), coal price, WP price and capacity factor. These variables are uncertain on their mean value and are available only on an upper and a lower bound of a range. Hence, these variables are assumed to uniform distribution as their probabilistic density function [196,197]. The nominal values of these variables reference the report from NETL [102] and the paper from Wei et al. [198].

Table 3-3 Statistical parameters of stochastic variables

Stochastic variable	Unit	Distribution	Nominal value	Range of distribution
Carbon tax	€/tCO ₂	Uniform	0	0-150
CO ₂ TS&M unit cost	€/tCO ₂	Uniform	9.73	5-15
Discount rate	%	Uniform	11.10	5-15
Oxygen carrier (Mn-ore) price	€/t	Uniform	100	50-200
coal price	€/GJ	Uniform	2.5	1-5
WP price	€/GJ	Uniform	4.5	3-15
capacity factor	%	Uniform	85	70-90

4 Oxy-fuel combustion power plants with the recuperated supercritical CO₂ power cycle

4.1 Process description

In this work, the reference case is a 660 MW_{el, gross} oxy-fuel CFPP developed by Hanak et al. [174]. Technical retrofits and improvements were considered based on the reference case to explore opportunities for reduction in efficiency and economic penalties. Section 4.1.1 and Section 4.1.2, respectively, define the detailed descriptions for the reference and retrofitted cases with key process assumptions shown in Table 4-1.

4.1.1 Reference oxy-fuel combustion coal-fired power plant with the supercritical steam cycle

The reference oxy-fuel combustion CFPP consists of a once-through boiler, a supercritical steam cycle with a single reheat and a flue gas treatment system. Under base-load operation, superheated steam of 537°C and 235 bar is raised in the oxy-fuel combustion boiler, which drives a high-pressure steam turbine with an outlet pressure of 45.2 bar. Steam is then reheated to 554°C in the boiler and subsequently drives the intermediate- and low-pressure steam turbines. The feedwater heating train comprises five low-pressure feedwater heaters and three high-pressure feedwater heaters to maximise steam cycle efficiency. The last low-pressure feedwater heater, called the deaerator, is a mixed feedwater heater.

It is assumed that O₂ is supplied at 95vol% purity from a cryogenic ASU with a specific power requirement of 200 kW_{el}/h/tO₂. Moreover, two-thirds of the flue gas from the oxy-combustion of coal is recycled to maintain combustion conditions in the chamber similar to that of the reference air-combustion case. After mixing with O₂ from the ASU, the concentration of O₂ in the mixed gas for coal combustion is fixed at 27vol%, which is higher than 21vol% of O₂ concentration in the air. The excess air and air leakage into the boiler are 20vol% and 2vol%, respectively. This results in a CO₂ concentration of more than 60vol% in the flue gas, with the

balance being H₂O and trace amounts of O₂, N₂, and Ar. The flue gas is then purified in the CPU which comprises a double-flash separation unit with internal cooling, to remove H₂O and the non-condensable species, thereby satisfying conditions for CO₂ transport and storage (110 bar and over 90vol% CO₂ purity [174]). To ensure a fair comparison between the cases analysed in this study, the work recovery system proposed by Hanak et al. [174] is not considered.

Table 4-1 Summary of the key process assumptions

Parameter		Value
Oxy-combustion coal-fired power plant		
Boiler	Design excess air ratio (vol%, dry) [174]	20.0
	Design air leakage (vol%, wet) [174]	2.0
	Discharge pressure of the induced draft fan (bar) [174]	1.05
	Discharge pressure of the forced draft fan (bar) [174]	1.11
	Isentropic efficiency of fans (%) [174]	75.0
	Mechanical efficiency of fans (%) [174]	100.0
	Recycled flue gas ratio (%) [174]	64.8
	Design O ₂ content in oxidising medium (vol%, wet) [174]	30.0
sCO ₂ cycle system	Initial turbine inlet temperature (°C) [199]	650
	Initial turbine outlet pressure (bar) [199]	250
	Isentropic efficiency of the sCO ₂ turbine (%) [199]	0.93
	Isentropic efficiency of the compressor (%) [199]	0.85
	Mechanical efficiency of the sCO ₂ turbine and the compressor (%) [199]	0.99
	Design discharge temperature of the cooler (°C) [199]	31.25
	Design discharge pressure of the cooler (bar) [199]	7.4
Auxiliary equipment		
Generator	Electrical efficiency (%) [199]	98.5
CO ₂ purification compression unit	Polytropic efficiency of CO ₂ compressors (%) [174]	79.0-80.0
	Mechanical efficiency of CO ₂ compressors (%) [174]	99.6
	Isentropic efficiency of CO ₂ pump (%) [174]	85.0
	Intercooling temperature (°C) [174]	40.0
	CO ₂ initial compression pressure (bar) [174]	30.0
	CO ₂ final pressure (bar) [174]	110.0
	CO ₂ temperature at inlet pump (°C) [174]	20.0
	ASU specific power (kW _{el} /tO ₂) [200]	200
Air separation unit	O ₂ purity (vol%) [174]	95.0
	Polytropic efficiency of air compressors (%) [174]	79.0-80.0
	Intercooling temperature (°C) [174]	40.0

4.1.2 Technical improvement

The retrofitted oxy-fuel combustion power plants (Figure 4-1 with the process flow diagram Figure 4-2) were modelled in Aspen Plus™ based on the oxy-fuel combustion boiler from the reference oxy-fuel CFPP. The thermodynamic properties were estimated using the Peng Robinson cubic equation of state with the Boston-Mathias alpha function (PR-BM) for the oxy-fuel combustion, ASU and CPU. The PR-BM was selected as it is recommended for hydrocarbon applications. Moreover, the Lee-Kesler-Plöcker equation of state (LK-PLOCK) was used to accurately represent the CO₂ properties under supercritical conditions in the sCO₂ power cycle. The main assumptions for the cases considered in this work are presented in Table 4-1.

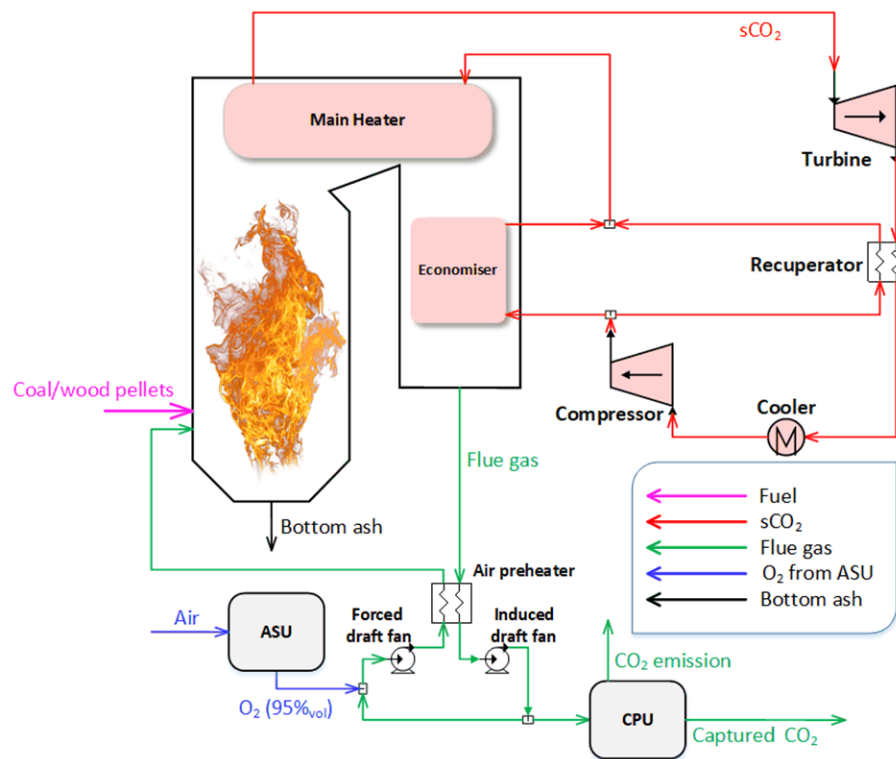


Figure 4-1 Improved oxy-fuel power plant flow diagram (including oxy-fuel combustion boiler, ASU, CPU and sCO₂ power cycle system) [198]

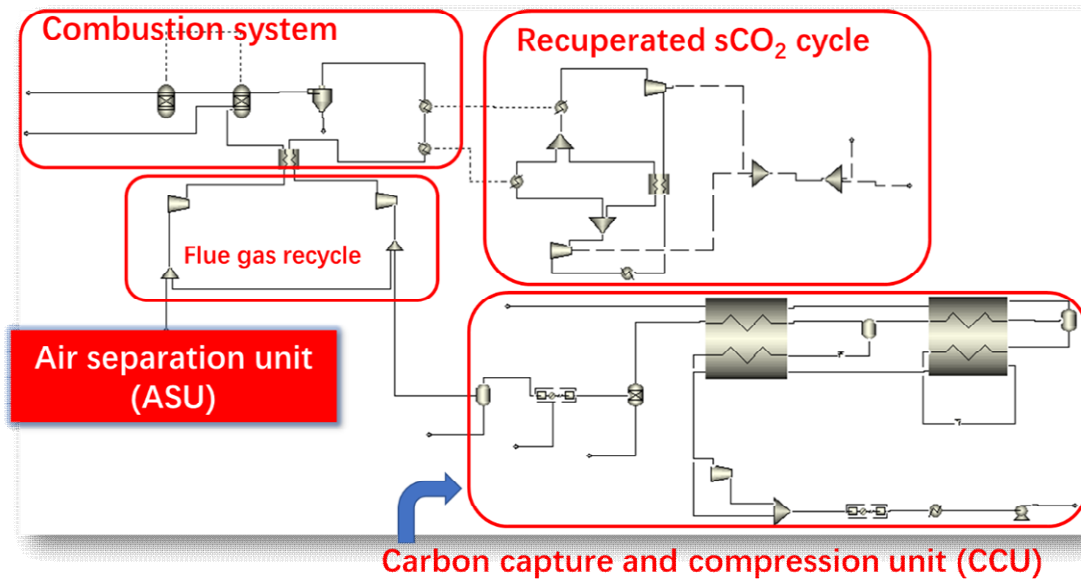


Figure 4-2 The process flow diagram of the proposed oxy-fuel combustion system with recuperated sCO₂ power cycle (only the ASU is considered as a black box.)

The sCO₂ cycle is a closed recuperated sCO₂ Brayton cycle with one sCO₂ turbine, one compressor and one recuperator. In the integrated system, the low-temperature, low-pressure CO₂ is compressed to 250 bar in the compressor. It is then divided into two streams that are preheated by two different heat sources. The first stream is preheated by the flue gas stream at the exit of the boiler. The second stream is preheated by the CO₂ stream discharged from the sCO₂ turbine. Both streams are then mixed and further heated in the oxy-fuel combustion boiler to 650°C [199], as assumed in Table 4-1 Summary of the key process assumptions. The high-pressure, high-temperature sCO₂ stream is then expanded in the turbine, which subsequently drives the generator to produce power. As a result, the temperature and pressure of the sCO₂ stream leaving the turbine are about 519°C and 77 bar, respectively. These are further reduced to 31°C and 74 bar, above the critical point for CO₂, in the recuperator and the cooler. The solid fuels considered in this work are coal (South African bituminous coal) [189] and biomass (wood pellet, WP) [190] with the compositions presented in Table 4-2. Based on the above considerations, the following cases are further investigated:

- SC-coal case – Oxy-fuel combustion CFPP with a supercritical steam cycle.
- sCO₂-coal case – Oxy-fuel combustion CFPP with a recuperated sCO₂ cycle.
- sCO₂-WP case – Oxy-fuel combustion biomass-fired power plant with a recuperated sCO₂ cycle.

Table 4-2 Fuel composition

Parameter	Coal [189]	Wood pellets [190]
Ultimate analysis		
Carbon (%wt, dry basis)	69.80	46.96
Hydrogen (%wt, dry basis)	3.58	6.86
Nitrogen (%wt, dry basis)	1.73	0.26
Oxygen (%wt, dry basis)	7.66	44.43
Chlorine (%wt, dry basis)	0	0
Sulphur (%wt, dry basis)	0.55	0.21
Ash (%wt, dry basis)	16.68	1.28
Proximate analysis		
Ash (%wt, dry basis)	16.68	1.28
Volatiles (%wt, dry basis)	22.85	85.00
Fixed carbon (%wt, dry basis)	60.47	13.72
Moisture (%wt)	2.44	5.34
Higher heating value (MJ/kg)	26.74	17.02

4.2 Consideration of sensitivity analysis

In the TEA, the net efficiency of the power plant, the specific CO₂ emissions, LCOE and AC were considered as the key performance indicators. Furthermore, identification of the optimum parameters of the working medium is potentially one way to improve the net efficiency and reduce the costs associated with CCS, such as raising the TIT and the COP [201]. Moreover, the economic performance depends upon the assumptions made in the analysis. It is pertinent to assess the effect of the carbon tax, fuel price, and discount rate on LCOE and AC. Therefore, this work analysed the effect of these parameters on the techno-economic performance of the cases identified above. The initial working medium parameters in the sCO₂ cycle cases are 650°C and 250 bar [199], and the remaining assumptions are shown in Table 3-1 and Table 4-2. The sensitivity analysis was performed to evaluate the effect of TIT, COP, carbon tax, fuel price, and discount rate on the techno-economic performance of the considered cases by varying:

- TIT between 500–900°C [202];
- COP between 100–300 bar;
- Carbon tax between 0–140 €/t;
- Fuel price by $\pm 25\%$; and
- Discount rate between 1.0–15.0%.

A technical report from Sandia National Laboratories shows that the TIT is in the range 550-900°C. The minimum temperature requirement for the primary heat exchanger producing sCO₂ is 550°C and the higher depends on the application [202]. The lowest COP was selected as 100 bar just above the critical pressure of CO₂ and the highest was selected with a typical ultra-supercritical pressure of 300 bar. The highest carbon tax is selected at 140 €/t that is close to the historic high level, such as Sweden, and the 0 €/t means no carbon tax is implemented. The fuel price is an indeterminate quantity and varies according to the market, so choose a range of 25% above or below the assumed fuel price. The discount rate is assumed 11.1% in univariate analysis. In the sensitivity analysis, the discount rate fluctuates by nearly 5%.

4.3 Thermodynamic assessment

The analysis of the thermodynamic performance (Table 4-3) revealed that the net efficiency of the SC-coal case decreased by 10.25%_{HHV} points, with respect to the conventional CFPP with a net efficiency of 38.80%_{HHV}, validated by Hanak et al. [174]. The net efficiency in the sCO₂-coal and the sCO₂-WP cases decreased by 10.70%_{HHV} points and 13.02%_{HHV} points, respectively. In these cases, the net power output decreased by 7.33 MW_{el} and 45.22 MW_{el}, respectively, with respect to the SC-coal case. This corresponds to energy penalties of 1.57%_{HHV} and 9.70%_{HHV}. It needs to be highlighted that the considered structure of the sCO₂ cycle is significantly less complex than that of the conventional steam cycle in the reference case. Its operating conditions need to be further optimised to improve the net efficiency of the sCO₂-coal case. However, a higher net efficiency penalty in the sCO₂-WP case is primarily due to the fuel characteristics of biomass. As a

result, optimisation of the sCO₂ cycle conditions may not result in a significant reduction of the net efficiency penalty in the sCO₂-WP case.

Table 4-3 Summary of the thermodynamic performance

Parameter	Unit	SC-coal case	sCO ₂ -coal case	sCO ₂ -WP case
Chemical energy consumption	MW _{th}	1632.97	1632.97	1632.97
Boiler heat-exchange efficiency	% _{HHV}	89.56	92.45	79.99
Auxiliary electricity requirements				
Condensate pump	MW _{el}	0.63	0.00	0.00
O ₂ -rich stream compressor (the forced draft fan)	MW _{el}	1.49	3.73	4.05
Flue gas booster (the induced draft fan)	MW _{el}	5.27	7.11	6.69
Coal/Wood Pellets handling and pre-treatment, flue gas treatment trains	MW _{el}	11.25	11.39	18.13
Cooling tower pump	MW _{el}	2.21	2.29	2.18
Plant auxiliary equipment electricity consumption	MW _{el}	20.85	24.52	31.05
Plant control systems, lighting, HVAC*, steam turbine auxiliaries, transformer losses	MW _{el}	5.00	4.95	4.71
ASU power consumption	MW _{el}	94.06	89.38	89.38
CPU power consumption	MW _{el}	73.37	74.62	75.67
System performance indicators				
Gross power output	MW _{el}	659.45	652.26	621.77
Total auxiliary electricity consumption	MW _{el}	193.28	193.46	200.81
Net power output	MW _{el}	466.17	458.79	420.96
Gross electric efficiency	% _{HHV}	40.38	39.94	38.08
Net electric efficiency	% _{HHV}	28.55	28.10	25.78
Specific fuel consumption	g/kW _{el} h	465.75	479.14	830.99
CO ₂ emission rate	kg/s	11.73	11.40	-146.78
Annual CO ₂ emission rate	MtCO ₂ /a**	0.30	0.29	-3.70
Specific CO ₂ emission	gCO ₂ /kW _{el} h	90.61	89.49	-1255.21
Net efficiency penalty	% _{HHV}	10.25	10.70	13.02

*HVAC: Heating, Ventilation and Air Conditioning

** Refers to CO₂ emission rate per annum

An analysis of the distribution of the auxiliary power consumption (Figure 4-3) has indicated that the ASU power consumption is the primary source of the efficiency penalty in all considered cases. In the sCO₂-coal case and the sCO₂-WP case, the ASU power consumption accounted for 13.7% and 14.4% of the gross power output, respectively. This corresponds to 46.2% and 44.5% of the auxiliary power consumption, respectively, and is in line with the results reported in the literature

[203]. In addition, the CPU power requirement in these cases amounted to 38.6% and 37.7%, respectively.

Furthermore, an analysis of the specific fuel consumption revealed that its value in the sCO₂-WP case (830.99 g/kW_{el}h) is about 1.75 times that of the other two cases. This can be associated with the lower HHV of the WP (16.8 MJ/kg) compared to that of coal (26.7 MJ/kg). Moreover, the specific CO₂ emissions were comparable in the SC-coal case (90.61 gCO₂/kW_{el}h) and the sCO₂-coal case (89.48 gCO₂/kW_{el}h), while the sCO₂-WP case led to a negative CO₂ emission (-1255.21 gCO₂/kW_{el}h) that corresponds to an atmospheric CO₂ removal rate of 3.70 MtCO₂ per annum. As a result, the sCO₂-WP case could offset CO₂ emissions from other processes with a potential benefit of an additional revenue stream from the ETS.

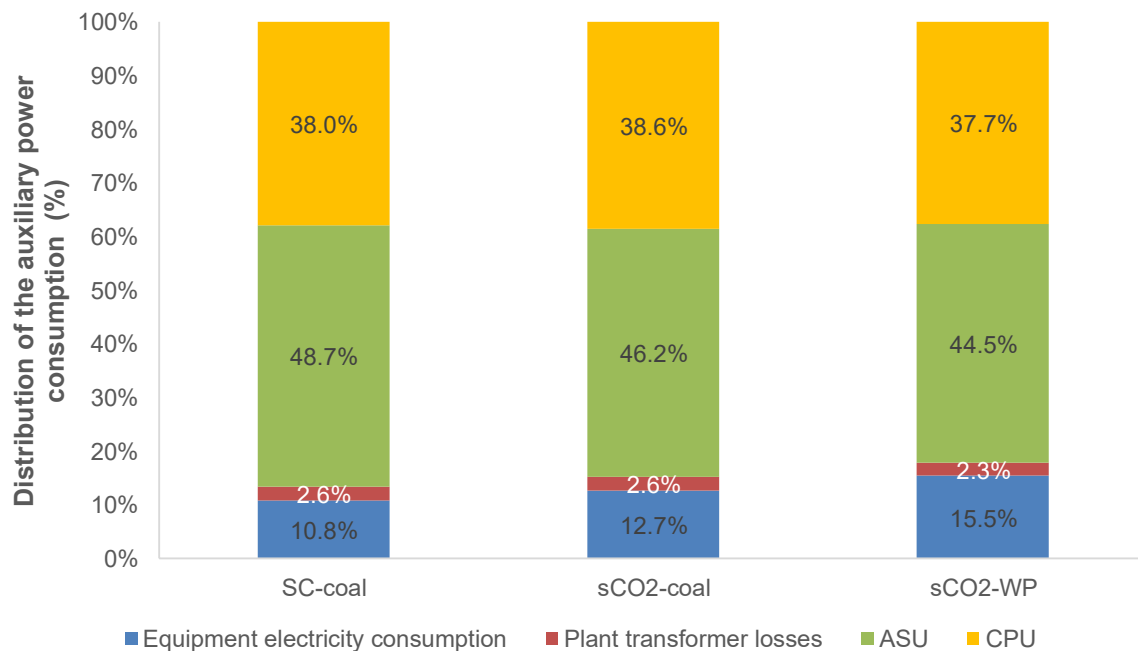
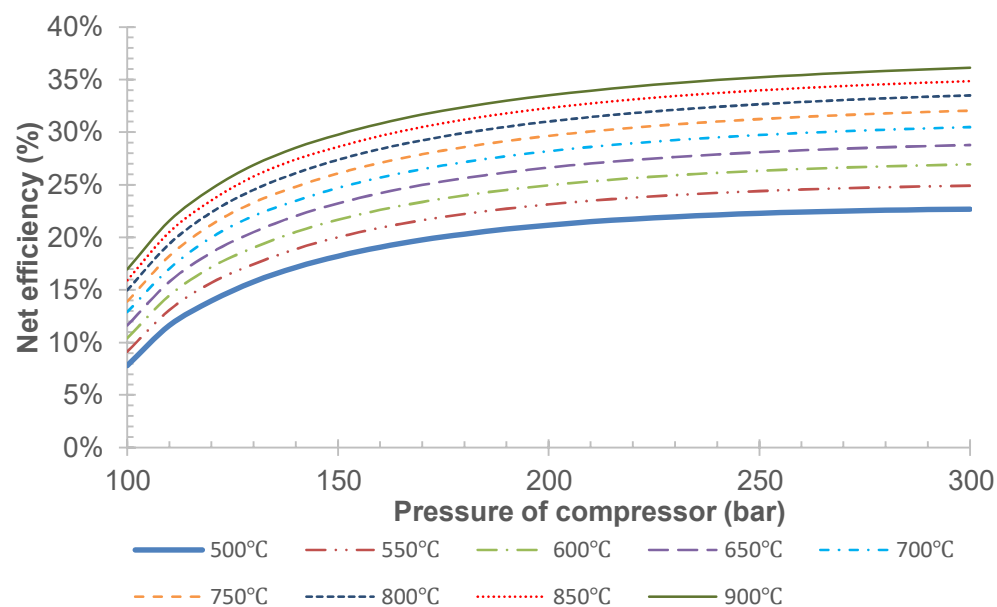


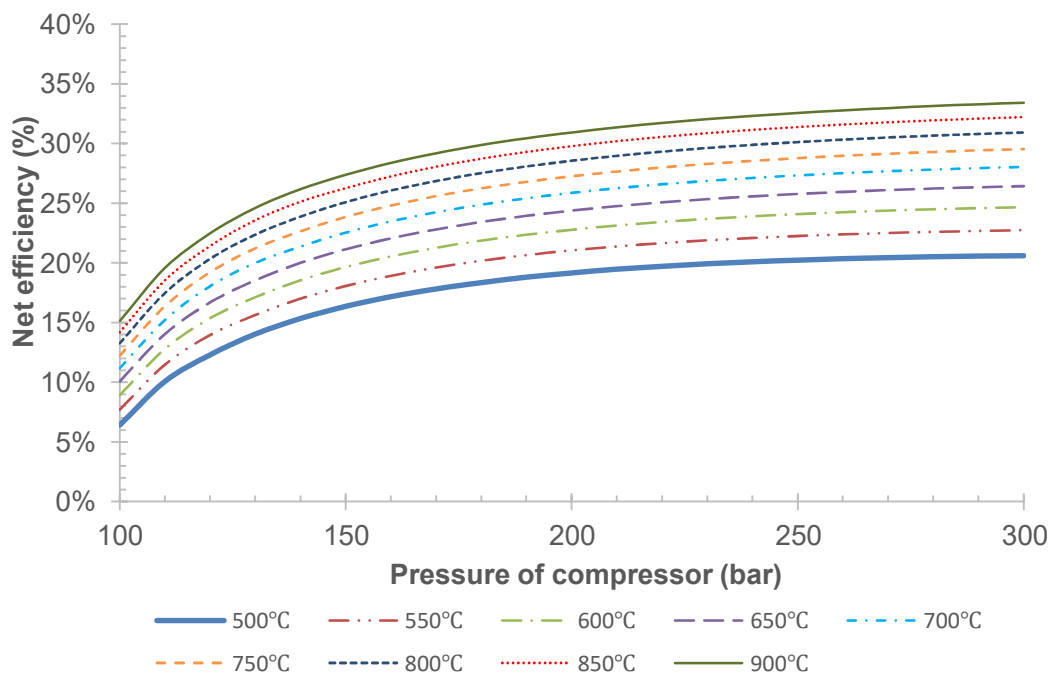
Figure 4-3 Distribution of the auxiliary power consumption

As mentioned above, the poorer performance of the cases with the sCO₂ cycle can be a result of non-optimised operating conditions. Figure 4-4 shows a positive correlation between net efficiency and both the TIT and the COP. As the COP increases from 250 bar to 300 bar at a TIT of 650°C, the net efficiency has

increased from 28.10%_{HHV} to 28.78%_{HHV} in the sCO₂-coal case and from 25.78%_{HHV} to 26.43%_{HHV} in the sCO₂-WP case. Under a fixed amount of heat released from the oxy-combustion of fuel, the flow rate of the sCO₂ is reduced as the COP increases. Consequently, the energy consumption of the compressor and the amount of heat rejected in the cooler are reduced. These led to an increase in the net efficiency of the sCO₂ cases. However, the net efficiency gains are significantly smaller at pressures above 250 bar. This can be explained by an increase in the CO₂ density from 771.5 kg/m³ at 100 bar to 922.4 kg/m³ at 250 bar (~1 kg/(m³*bar) increase) and 948.0 kg/m³ at 300 bar (~0.5 kg/(m³*bar) increase). As a result, the increase in the energy requirement of the main compressor became close to the gains in the work output of the expander at higher pressures, resulting in lower gains in the net power output. Furthermore, Figure 4-4 reveals that an increase in the TIT from 650°C to 750°C, at a constant COP of 250 bar, increased the net efficiency from 28.10%_{HHV} to 31.25%_{HHV} in the sCO₂-coal case and from 25.78%_{HHV} to 28.78%_{HHV} in the sCO₂-WP case. Therefore, the sCO₂ cycle cases have the potential to offer superior net efficiency to the conventional steam cycle (28.55%_{HHV}) at higher TIT and COP values. It needs to be highlighted that, to maximise the net efficiency, the TIT should be as high as possible, considering material limitations.



(a)



(b)

Figure 4-4 Effect of the turbine inlet temperature and the pressure of compressor on the net efficiency in a) sCO₂-coal case and b) sCO₂-WP case

4.4 Economic assessment

The initial economic evaluation of the considered cases (Table 4-4) indicated that replacement of the conventional steam cycle (SC-coal case) with the sCO₂ cycle (sCO₂-coal case) resulted in a TCR reduction of 20.1 M€. This was because of the lower capital cost of the sCO₂ cycle. This is reflected in a 4.1 €/kW_{el, net} reduction in the specific capital cost in the sCO₂-coal case. Importantly, on a change of fuel from coal (sCO₂-coal case) to WP (sCO₂-WP case), the TCR was 57.4 M€ lower than that in the reference case. However, the specific capital cost increased by 130.4 €/kW_{el, net}, due to the significantly lower net efficiency of the sCO₂-WP case compared to the SC-coal case.

In Figure 4-5, it shows the distribution of LCOE under initial assumptions. The economic performance assessment has also revealed that, compared to the LCOE of the SC-coal case (88.3 €/MW_{el,h}), the values for the sCO₂-coal case and the sCO₂-WP case have increased by 0.6 €/MW_{el,h} (0.7%) and 31.8 €/MW_{el,h} (36.0%), respectively. Such an increase in the latter case is because of a 60.7% higher price for WP (4.5 €/GJ) compared to that of coal (2.8 €/GJ) [97] and lower net efficiency in the sCO₂-WP case shown in Table 4-4. As a result, the fuel component of the LCOE in the sCO₂-WP case is 76.9% higher than that in the sCO₂-coal case. Therefore, when the sCO₂-WP case is not considered to receive benefits from CO₂ offsetting, it is more economical to use coal rather than the WP as fuel.

In Table 4-4, the sCO₂-WP case showed significantly higher LCOE with 31.2 €/kW_{el,h} compared with the sCO₂-coal case. It because of the 2.3%_{HHV} of net efficiency penalty resulting from the lower heating value and higher moisture of biomass. Even so, analysis of the AC presented more favourable results that support BECCS deployment. Namely, the AC for the sCO₂-WP case was estimated to be 38.5 €/tCO₂, 41.1% lower than that of the SC-coal case (65.3 €/tCO₂). On the contrary, the AC of the sCO₂-coal case was higher than the SC-coal case by 0.7 €/tCO₂ (1.1%). This can be explained primarily by negative CO₂ emissions in the sCO₂-WP case (-1255.21 gCO₂/kW_{el,h}) compared to still positive

CO₂ emissions in the other cases (~90 gCO₂/kW_{el}h). It can be expected, therefore, that BECCS may become favoured economically over the CCS systems considering the potential revenue from ETS, even though their net efficiencies are lower than that of CCS.

Table 4-4 Summary of economic performance

Parameter	Unit	SC-coal case	sCO ₂ -coal case	sCO ₂ -WP case
Gross power output	MW _{el}	659.45	652.26	621.77
Net power output	MW _{el}	466.17	458.79	420.96
Net electric efficiency	% _{HHV}	28.55	28.10	25.78
Fuel price	€/GJ	2.8	2.8	4.5
Total capital requirement	M€	1157.8	1137.7	1100.4
Specific capital cost	€/kW _{el, net}	2483.6	2479.5	2614.0
Annual fuel cost	M€/a	114.2	114.2	185.4
Annual fixed operating cost	M€/a	11.6	11.4	11.0
Annual variable operating cost	M€/a	47.0	46.6	47.9
Levelised cost of electricity	€/MW _{el} h	88.3	88.9	120.1
CO ₂ avoided cost	€/tCO ₂	65.3	66.0	38.5

The results presented in Figure 4-6 and Figure 4-7 revealed that both the COP and the TIT are negatively correlated with the LCOE and the AC for coal and WP fuels. This implies that an increase in both parameters will improve the economic performance of the considered cases. The trend could be correlated with the influence of the COP and the TIT on the net efficiency of considered cases, as presented in Figure 4-4. This means that the more efficient the power plant, the lower the LCOE and the lower the amount of emitted CO₂ when generating a kilowatt of electricity (e_{CO_2}). However, the largest reduction in both LCOE and AC are achieved at the lower end of the ranges considered for the COP and the TIT, whereas improvements above 250 bar and 700°C are relatively marginal.

Furthermore, due to a higher price of WP compared to that of coal, the LCOE in the sCO₂-WP case is significantly higher than that of the sCO₂-coal case (Figure 4-6). This, subsequently, resulted in relatively poor performance of the sCO₂-WP case in the sensitivity analysis (Figure 4-6b). However, because the former case can achieve negative CO₂ emissions, a substantial difference in the AC was observed between the sCO₂-coal (Figure 4-7a) and the sCO₂-WP (Figure 4-7b)

cases. This confirms that power plants based on BECCS can become more economically feasible than the CCS systems, achieving a competitive advantage in the market driven by carbon tax prices.

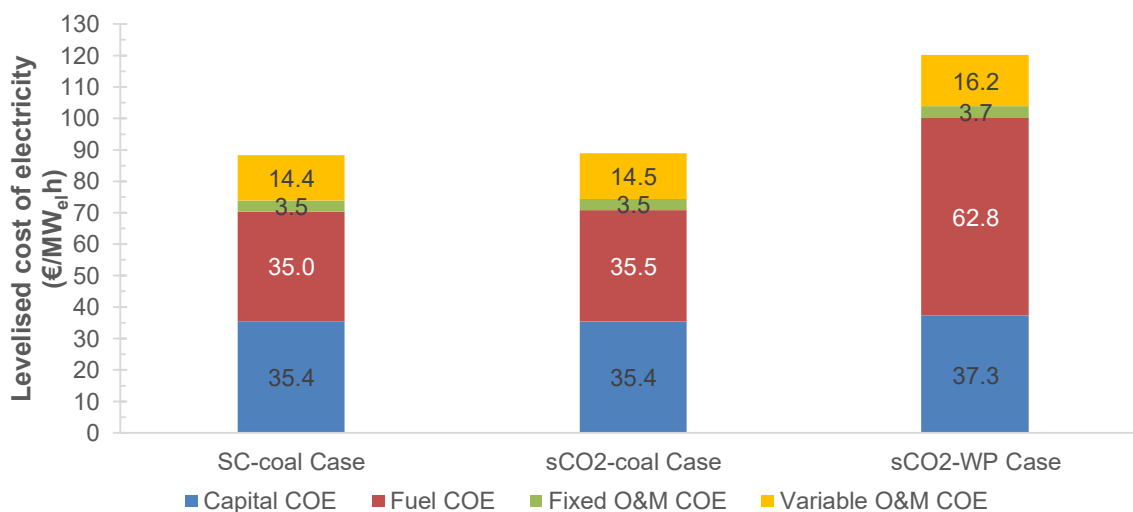
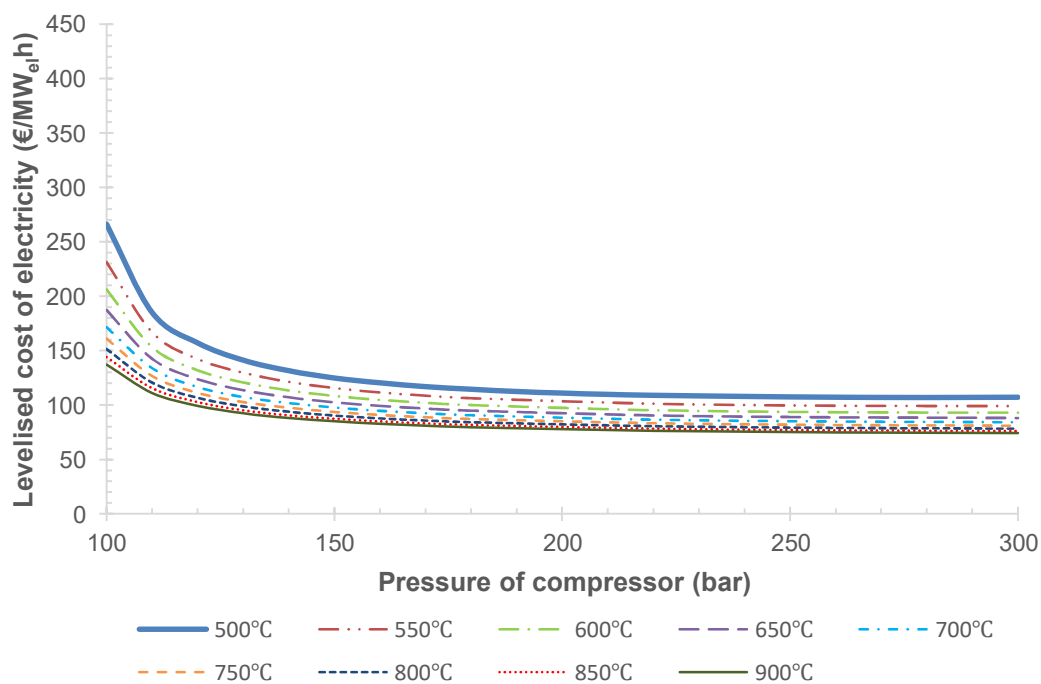
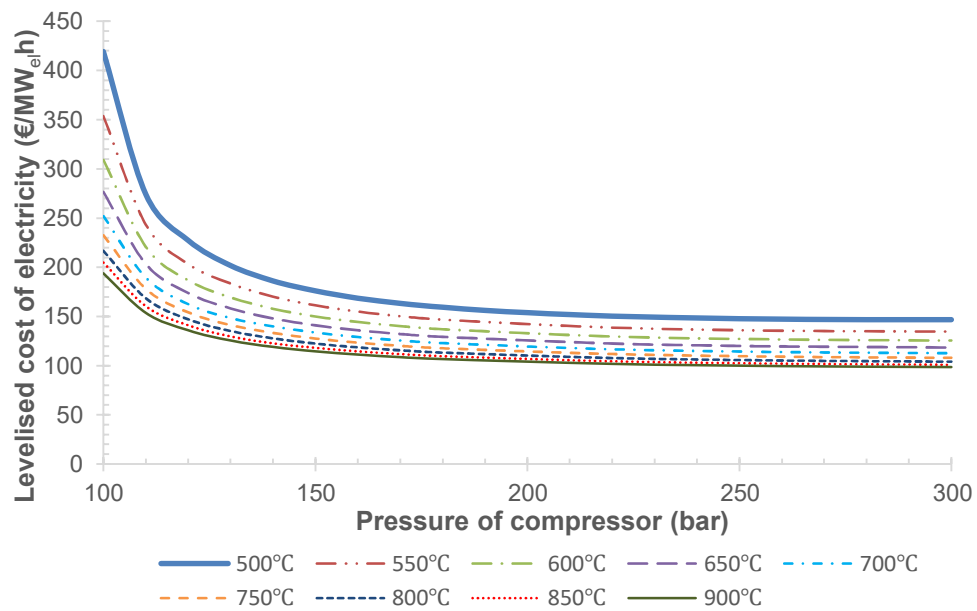


Figure 4-5 Distribution of levelised cost of electricity under initial assumptions

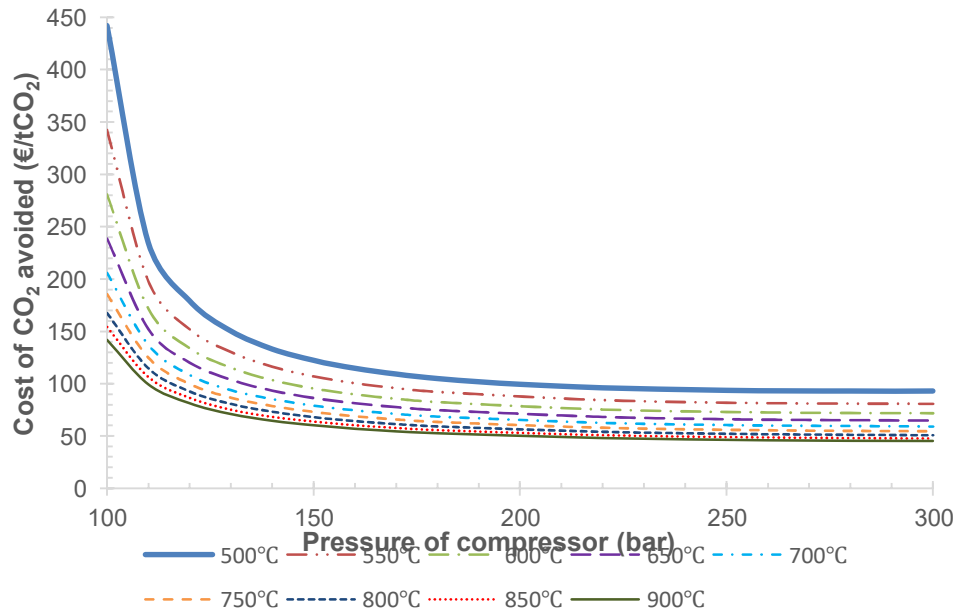


(a)

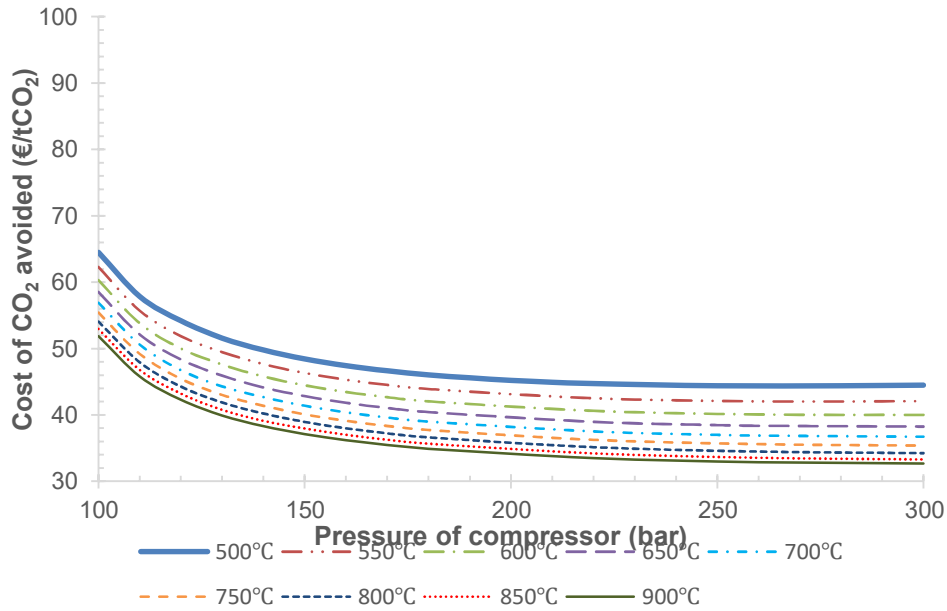


(b)

Figure 4-6 Effect of the turbine inlet temperature and the pressure of compressor on the levelised cost of electricity in a) sCO₂-coal case and b) sCO₂-WP case



(a)



(b)

Figure 4-7 Effect of the turbine inlet temperature and the pressure of compressor on the cost of CO₂ avoided in a) oxy-fuel combustion coal-fired power plant with sCO₂ cycle and b) oxy-fuel combustion biomass-fired power plant with sCO₂ cycle

To understand the benefits of BECCS over CCS in the market driven by the carbon tax, the effect of the carbon tax on the economic performance of sCO₂-coal and sCO₂-WP cases has been evaluated (Figure 4-8). This analysis has revealed that the parity of LCOE will be achieved for both cases at the carbon tax of 23.18 €/tCO₂. Such a result indicates that when the carbon tax is more than 23.18 €/tCO₂, the sCO₂-WP case is more economically favoured. Importantly, with the current CO₂ price of the European emission allowance of 23.74 €/tCO₂ [204], the sCO₂-WP case would be a more economically feasible option. Moreover, if the carbon tax is 95.70 €/tCO₂, the revenue generated by offsetting CO₂ emissions in the sCO₂-WP results in an LCOE of zero.

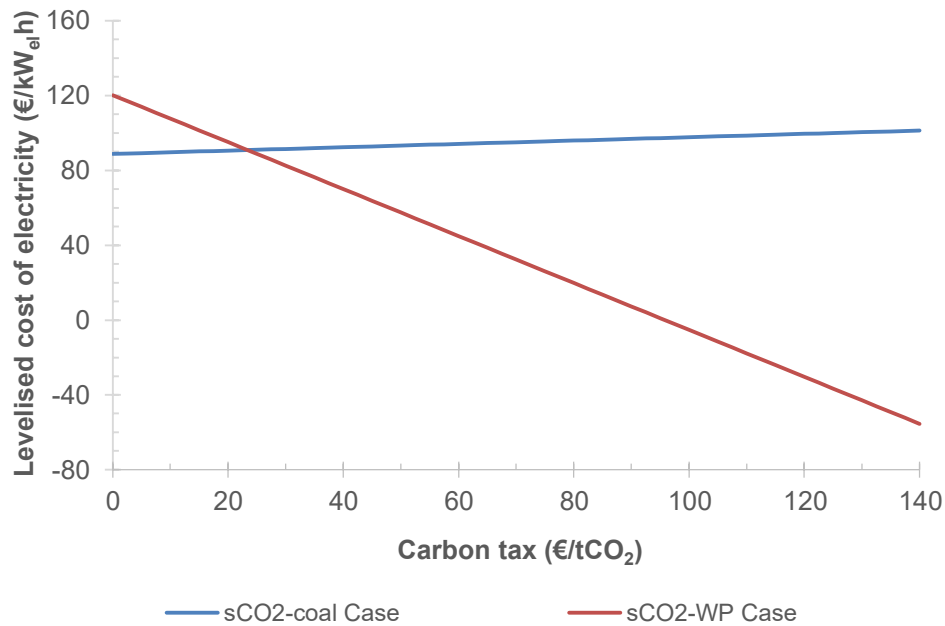


Figure 4-8 Effect of the carbon tax on the levelised cost of electricity

Furthermore, it is pertinent to assess the effect of the economic assumptions made in Table 3-1 on the economic performance of the considered cases. Figure 4-9 reveals that the fuel price significantly influences both the LCOE and the AC. On a 25% increase in the fuel price, the LCOE of the sCO₂-WP case increased by 35.4% (141.1 €/MW_eh), a significantly higher increase than that in the sCO₂-coal case, for which the LCOE has increased by 17.8% (104.7 €/MW_eh). Importantly, even if the price of WP and coal are comparable, between 3.0–4.0 €/GJ, the LCOE of the sCO₂-WP case was found to be higher than that of the sCO₂-coal case by 7.3–8.4 €/MW_eh. This was primarily due to the lower net efficiency of the sCO₂-WP case. Furthermore, even though the price of coal is lower, the AC of the sCO₂-coal case is much higher than that of the sCO₂-WP case (Figure 4-9). This is due to negative CO₂ emissions in the sCO₂-WP case. Moreover, a variation in the coal price has a greater impact on the AC than that in the WP price. This indicates a potentially higher resilience of BECCS to uncertainty in the future energy market.

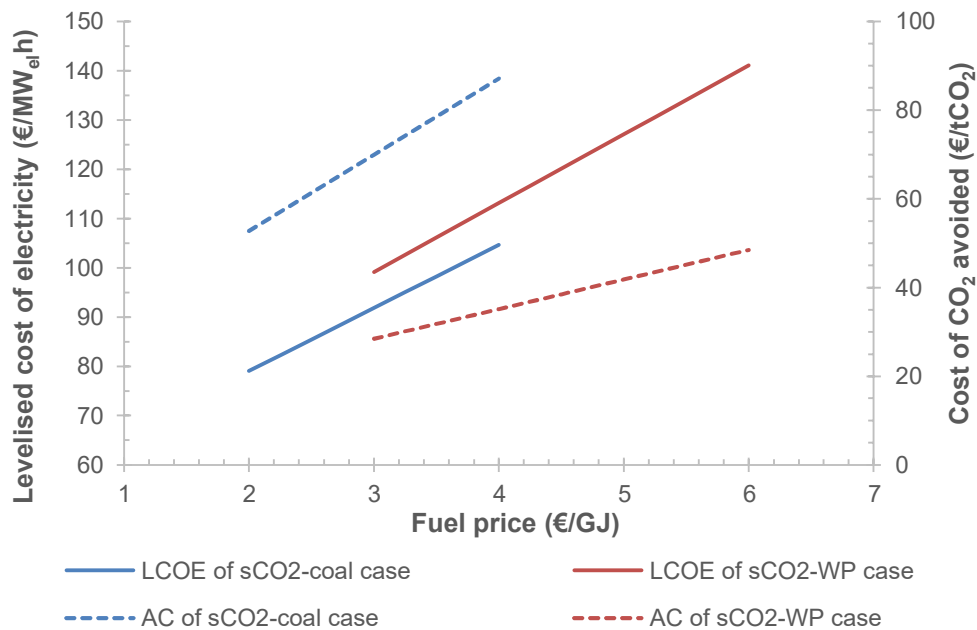


Figure 4-9 The effect of the fuel price on the levelised cost of electricity and the cost of CO₂ avoided

Finally, the results presented above were determined based on a discount rate of 8.78% (Table 3-1). As this figure depends on the level of risk associated with the project and directly influences the considered economic performance indicators, it is pertinent to assess its impact on the economic feasibility of the considered cases. Figure 4-10 revealed that an increase in the discount rate will result in a subsequent increase in the LCOE. Importantly, the observed trend is comparable for both sCO₂-coal and sCO₂-WP cases. On the contrary, Figure 4-10 indicated that the AC in the former case is more sensitive to an increase in the discount rate, with rather low sensitivity in the sCO₂-WP case. This indicates that the BECCS systems may be less sensitive to the risks associated with the project delivery.

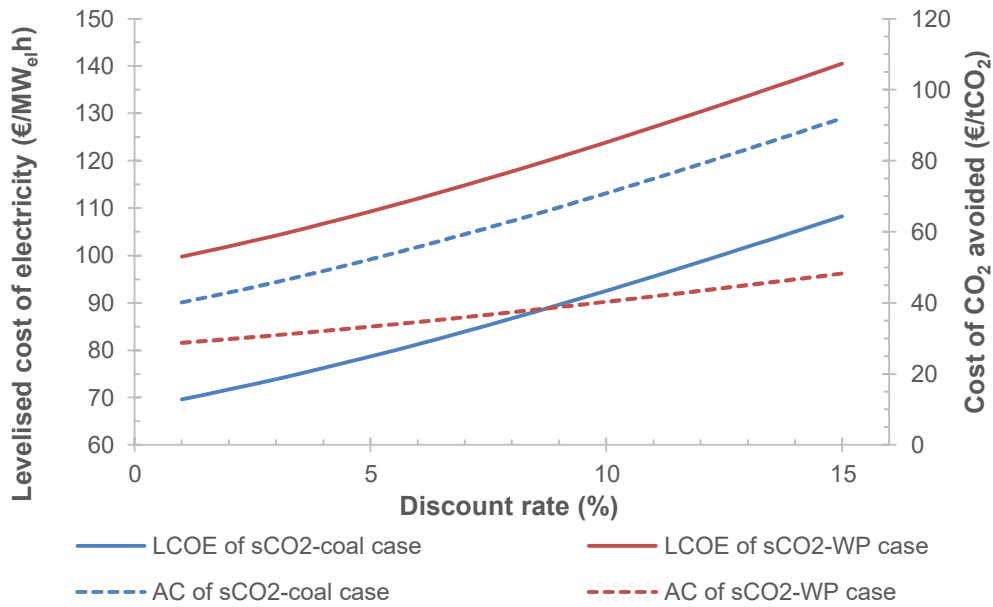


Figure 4-10 The effect of the discount rate on the levelised cost of electricity and the cost of CO₂ avoided

4.5 Feasibility assessment

Considering the outcome of the initial TEA and corresponding sensitivity analyses, the potential for further improvement in the performance of the oxy-fuel combustion power plant with the sCO₂ cycle has been indicated. Therefore, the techno-economic feasibility of the considered cases has been evaluated under the revised conditions (Table 4-5), considering the outputs from the sensitivity analyses. These indicated that substantial improvements in the economic performance occurred until the TIT and COP reached 700°C and 250 bar, respectively. Under such operating conditions, the net efficiency of both the sCO₂-coal and sCO₂-WP case increased by around 1.6%_{HHV} points. Compared to the SC-coal case, the net efficiency under revised conditions was 1.2%_{HHV} points higher for the sCO₂-coal case and 0.5%_{HHV} points lower for the sCO₂-WP case. An increase in the COP to 300 bar resulted in a further gain in the net efficiency of 0.8%_{HHV} points (sCO₂-coal case) and 0.7%_{HHV} points (sCO₂-WP case). Overall, an increase in the operating conditions has reduced the efficiency penalty to 8.3%_{HHV} points (sCO₂-coal case)

and 10.8%_{HHV} points (sCO₂-WP case) with respect to the CFPP without CO₂ capture. This implies that the former case is superior to the reference SC-coal case (10.3%_{HHV} points), but the thermodynamic performance of the latter is still inferior.

Table 4-5 Summary of the techno-economic assessment

Parameter	Unit	SC-coal case	sCO ₂ -coal case				sCO ₂ -WP case		
Turbine inlet temperature	°C	537	650	700	700	650	700	700	
Compressor outlet pressure	bar	235	250	250	300	250	250	300	
Gross power output	MW _{el}	659.4	652.3	679.1	691.4	621.8	647.3	659.0	
Net power output	MW _{el}	466.17	458.8	485.5	497.8	421.0	446.4	458.0	
Total capital requirement	M€	1157.8	1137.7	1164.7	1196.1	1100.4	1127.4	1157.7	
Specific capital cost	€/kW _{net}	2483.6	2479.5	2398.8	2403.0	2614.0	2525.6	2527.6	
CO ₂ emission rate	kg/s	11.7	11.4	11.4	11.4	-146.8	-146.8	-146.8	
	MtCO ₂ /a	0.30	0.29	0.29	0.29	-3.70	-3.70	-3.70	
η_{net}	% _{HHV}	28.55	28.10	29.73	30.48	25.78	27.34	28.05	
η_{gross}	% _{HHV}	40.38	39.95	41.59	42.34	38.08	39.64	40.36	
Net efficiency penalty	% _{HHV}	10.3	10.7	9.1	8.3	13.0	11.5	10.8	
LCOE	€/MW _{el} h	88.3	88.9	85.1	84.2	120.1	114.4	112.7	
AC	€/tCO ₂	65.3	66.0	60.5	59.1	38.5	37.0	36.7	

From the economic feasibility perspective, an increase in the TIT and COP to 700°C and 250 bar, respectively, resulted in significant improvements in the economic performance of the sCO₂-coal case (Table 4-5). Namely, the LCOE reduced from 88.9 €/MW_{el}h to 85.1 €/MW_{el}h and the AC reduced from 66.0 €/tCO₂ to 60.5 €/tCO₂, both of which are below the figures reported for the SC-coal case. Further reductions by 0.9 €/MW_{el}h and 1.4 €/tCO₂, respectively, were obtained on an increase of the COP to 300 bar. In addition, some improvement in the economic performance of the sCO₂-WP case was also observed. However, this improvement was less pronounced than that in the sCO₂-coal case. This is because although a 4.7% reduction in the LCOE was achieved on an increase in the TIT and COP to 700°C and 250 bar, it is still substantially higher (114.4 €/MW_{el}h) than that of the other considered cases (Table 4-5). Although the LCOE in the sCO₂-WP case was 33-35% higher than that for the sCO₂-coal case, this increase is lower than that reported by Al-Qayim et al. [70] for oxy-fuel combustion power plants with a

conventional steam cycle. It also needs to be emphasised that even though the sCO₂-WP case was characterised with the lowest net efficiency, it had the lowest AC among the considered cases. Therefore, as discussed above, the main benefit of BECCS that makes it economically feasible relies on its inherent ability to operate with negative CO₂ emissions, subsequently offsetting CO₂ emissions from other sources that are difficult to decarbonise, such as the aviation or transportation sectors. Nevertheless, without the revenue from the carbon tax, the relatively poor techno-economic performance of the sCO₂-WP case compared to the other cases would not support the commercialisation of BECCS.

5 Chemical looping combustion power plants with the recompression supercritical CO₂ power cycle

5.1 Process description

Two cases in a report from Stevens et al. [178] are taken as the reference and further developed in the present study. Section 5.1.1 introduces the overall reference cases, and Section 5.1.2 introduces the improvements on process based on the reference cases.

5.1.1 Reference chemical looping combustion coal-fired power plant with the supercritical steam cycle

The reference case considered a new design of a 550 MW_{el,net} supercritical coal direct chemical looping combustion (CDCLC) power plant firing Illinois No. 6 (bituminous coal). The CDCLC power plant employs common plant site and environmental conditions in the American Midwest with operating conditions defined in the *Quality Guidelines for Energy System Studies: Process Modelling Design Parameters* [205]. It was designed that the steam conditions are supercritical (593°C/593°C, 240bar), with over 90% of carbon capture efficiency, over 95mol% of CO₂ product purity, and 153 bar of CO₂ product delivery pressure. For the cost estimation basis, the plant capacity factor is 85%. The high-risk financial assumption was used with the capital charge factor higher than 12.43%. The potential risks in this project include uncertainties in the technical scope, budget management, resources and schedule.

The heart of the power plant is the FR and AR, which are the CFBs. This study considers Fe₂O₃ and CaSO₄ as oxygen carriers that are circulated between FR and AR. The fluidized bed reactor model references the literature from Abba, et al [206] and Kunii, D., and O. Levenspiel [207]. Coal is delivered to the base of the FR using conventional coal handling and feeding equipment. A stream of makeup OC (Fe₂O₃ or limestone) is also fed at the FR base. After air preheater, recycled steam and CO₂ from the flue gas provide reactants for coal devolatilization and

facilitate the fluidization, in the FR. So called recycled CO₂ is from the FR, which has been compressed for the first time through the CO₂ compression system. It mainly contains CO₂, probably, with some unconverted fuel components. Key operation parameters for the reference CDCLC are listed in Table 5-1 and Table 5-2.

Table 5-1 Assumption of the fuel reactor and the air reactor characteristics in the reference plant

Parameter	Fe ₂ O ₃
<i>Fuel reactor</i>	
Temperature (°C) [178]	951.67
Pressure drop (bar) [178]	1.48
Excess oxygen carrier ratio (mass basis) [178]	1.56
Oxygen carrier makeup ratio (mass basis) [178]	0.00011
Oxygen carrier makeup rate (kg/s) [178]	1.3
Solids flow to Reducer (kg/s) [178]	11 890.92
Discharge pressure of the induced draft fan (bar) [178]	1.05
Discharge pressure of the gasification agent fan (bar) [178]	1.63
Polytrophic efficiency of gasification agent fans (%) [178]	75
Recycled enhancer gas/C in fuel (mass basis) [208]	4.17
<i>Air reactor</i>	
Temperature (°C) [178]	982.22
Pressure drop (bar) [178]	0.12
Design excess air ratio (vol%, dry) [178]	1.19
Off-gas H ₂ and CO (mol%) [178]	0.05
Oxygen carrier flow to Oxidizer (kg/s) [178]	11789.12
Off-gas O ₂ (mol%) [178]	3.5
Discharge pressure of the induced draft fan (bar) [178]	1.02
Discharge pressure of the forced draft fan (bar) [178]	1.23
Polytrophic efficiency of air fans (%) [174]	75

Table 5-2 Assumption of the recompression sCO₂ cycle system and the auxiliary equipment characteristics

Parameter	Value
<i>CLC system</i>	
Temperature of the fuel reactor and the air reactor (°C)	Depend on type of the OC, and assumed in Table 5-3
Pressure drop of flue gas through the preheater (bar) [116]	0.06
Pressure drop of depleted air through the preheater (bar) [116]	0.06
Discharge pressure of induced draft fans of flue gas (bar) [116]	1.01

Discharge pressure of induced draft fans of depleted air (bar) [116]	1.05
Polytropic efficiency of induced draft fans (%) [116]	75%
Discharge pressure of the gasification agent fan (bar) [116]	1.63
Polytropic efficiency of the gasification agent fan (bar) [116]	75%
Discharge pressure of the forced draft fan (bar) [116]	1.23
Polytropic efficiency of the forced draft fan (%) [116]	75%
Recompression sCO₂ cycle system	
Initial turbine inlet temperature (°C) [178]	593
Initial turbine outlet pressure (bar) [178]	240
Isentropic efficiency of the sCO ₂ turbine (%) [176]	93%
Isentropic efficiency of the main compressor (%) [176]	86%
Isentropic efficiency of the recompressor (%) [176]	86%
Mechanical efficiency of the sCO ₂ turbine and the compressor (%) [176]	99%
Design discharge temperature of the cooler (°C)	33
Design discharge pressure of the cooler (bar)	74
Pressure drop of heat exchangers HTR [39]	0.26
Hot side	0.09
Cold side	0.16
LTR [39]	0.08
Hot side	0.07
Cold side	0.72
Cooler [39]	0.35
Water wall [39]	
Superheater [39]	
Auxiliary equipment	
Electric motor efficiency (%) [178]	97%
Generator efficiency (%) [178]	98.5%
CO ₂ purification and compression units	
Compressor stage	6
Pressure drop in each stage (bar)	0.03
Polytropic efficiency of CO ₂ compressors (%) [174]	79.0-80.0%
Mechanical efficiency of CO ₂ compressors (%) [174]	99.6%
Isentropic efficiency of CO ₂ pump (%) [174]	85.0%
Intercooling temperature (°C) [174]	40.0
CO ₂ final pressure (bar) [174]	152.7

In the FR, the coal char, OC and coal ash particles entrained in the stream is captured in a set of four parallel cyclones, and then, are recycled to the bottom of the FR. A stream of entrained solids from the FR are transported to the AR through one of the cyclones. The flue gas from the FR mainly includes CO₂ and H₂O with portions of particulate, coal contaminants (SO₂, H₂S, HCl, Hg) and unconverted fuel constituents (primarily H₂ and CO), which is so called the raw CO₂ stream for

furthering separating and compressing. The particulate and coal contaminants have to be removed by conventional cleaning equipment. Regarding the unconverted fuel constituents, the OC based on Fe_2O_3 is sufficiently reactive in the FR resulting in the low H_2 and CO content in the FR's flue gas. Therefore, a fuel recovery unit is unnecessary, and conventional CO_2 stream dehydration and compression is applied. On the contrary, the relatively low reactivity of the CaSO_4 oxygen carrier results in a significant amount of H_2 and CO, and a fuel recovery/ CO_2 purification unit is applied to increase utilisation of the fuel and production of the high-purity CO_2 stream. A portion of the steam driving the power cycle system is produced by heat transfer with an HRSG before the gas cleanup equipment.

The FR is the most complex reactor in CLC with coal gasification and combustion. In the reaction gasifying coal, it is slow at the temperature of the FR, the unconverted char could be fed into the AR from FR with the OC circulation stream which results in the undesirable CO_2 emission. Therefore, a char and ash separation device is required for the FR solids transport stream, which could be design based on the mechanisms including density differences, size differences, and differences in magnetic properties. The plant LCOE has not been significantly impacted by the uncertainty of the separator's capital cost [178]. So, the cost of the separator is assumed to be 0 in this study. Then, it is recycled to the FR. The separation could reduce the volume of FR through increasing the content and residence time of char in the FR, and avoids unnecessary CO_2 emission.

The design of the AR is much simpler than that of the FR. In this reactor, the partially reduced OC from the FR is transported to the bottom of the AR and reacts with the air to produce almost completely oxidized OC. The AR uses eight parallel cyclones - six for recycling of entrained solids back into the AR and two for transporting a part of the entrained solids to the FR. The AR exhaust gas, named depleted air, mainly contains N_2 with excess O_2 , H_2O , particulates, CO_2 , NO_x and small amounts of coal contaminants. The baghouse and SCR are expected

enough for the gas cleaning process of AR exhaust gas. A conventional forced draft fan and conventional induced draft fans are utilized for airflow and flue gas, separately. The coal ash and OC waste are drained from the AR for maintaining a coal ash inventory balance within the reactors. The makeup OC is based on these losses of OC waste purged from AR and cyclone separation from off-gas of FR and AR.

Table 5-3 Basis oxygen carriers

	Fe ₂ O ₃	NiO	CaSO ₄	Mn ₃ O ₄	Fe ₂ TiO ₅
Oxygen carrier type [178]	<i>Manufactured material</i>			<i>Natural material</i>	
Makeup of oxygen carrier	Fe ₂ O ₃	NiO	Limestone [178]	Manganese ore	Ilmenite
Makeup carrier composition	45 wt% Fe ₂ O ₃ 55 wt% Al ₂ O ₃ [178]	32.7 wt% NiO 67.3 wt% Ni Al ₂ O ₄ [209]	100 wt% CaCO ₃	68.2 wt% Mn ₂ O ₃ 10.6 wt% Fe ₂ O ₃ [127]	29.2 wt% Fe ₂ O ₃ 63.9 wt% TiO ₂ [95]
Maximum oxygen transport capacity of pure OC system	0.0334 [115]	0.2142 [115]	0.4701 [115]	0.0509 [127]	0.05 [210]
Major redox pair	Fe ₂ O ₃ /Fe ₃ O ₄ [178]	NiO/Ni [115]	CaSO ₄ /CaS [178]	Mn ₃ O ₄ /MnO [127]	Fe ₂ TiO ₅ /FeTiO ₃ [115]
Attrition rate (wt%/h)	0.011% [178]	0.022% [115]	-	0.1%	0.4% [211]
Average lifetime of oxygen carrier (h)	9000	4500 [115]	200	1000 [127]	250 (100–300) [212]
Mole ratio of Ca to S	-	-	1 [178]	-	-
Char conversion efficiency (wt%)	96% [178]	92.8% [209]	96% [178]	97% [212]	96% [115]
Solids inventory (t/MW _{th})	0.05 (0.05–0.1) [115]	0.5 [115]		0.75 [212]	0.75 [212]
Oxygen carrier delivered price (€/t)	2000 (2000–9000) [178]	8000 [115]	33 (30–270) [178]	100 (100–400) [115]	100 (100–200) [212]

T_{FR}/T_{AR}	982°C/1093°C	950°C/1050°C	982°C/1093°C	950°C/1050°C	950°C/1000°C
[115,178]					

T_{FR} : fuel reactor temperature

T_{AR} : air reactor temperature

The SCR system is designed for 65% NO_x reduction using ammonia to reduce NO_x to N₂ and H₂O in a fixed catalyst bed with less than 2 ppmv ammonia slip. Then a vertical-shaft, tri-sector, regenerative air heater cools the flue gas to 143°C by preheating the combustion air. After that, a spray dryer absorber uses the lime slurry to remove SO₂ in the flue gas with 93% of the designed SO₂ removal efficiency. The following baghouse removes the remaining particulate matter.

Regarding the OCs (Table 5-3) applied in the reference CLC power plant, Fe₂O₃ supported on alumina (Al₂O₃) was considered, characterizing highly reactive, but expensive with the delivered price of 2,000-10,000 €/t. On the contrary, the CaSO₄ oxygen carrier used the low-cost limestone with the price of 90-270 € per ton. The actual mass flow rate of OC circulation should be higher than the theoretical mass flow rate of that, due to the maximum oxygen transport capacity was considered [115]. The losses of the Fe₂O₃ oxygen carrier are due to attrition which is used to estimate the OC makeup rate. In the case of CaSO₄, the mole ratio of Ca to S is used to estimate the limestone makeup flow rate.

5.1.2 Technical improvement

The following improvements are considered in this study with the proposed process shown as Figure 5-1 with the process flow diagram Figure 5-2:

- 1) The steam Rankine cycle is replaced by the recompression sCO₂ power cycle.
- 2) Additional flue gas recycles to fuel reactor: Based on the reference CLC power plant, fluidization with recycled flue gas from the fuel reactor was additionally considered to enhance the char gasification. The recycled flue gas is withdrawn before condensation containing around 96 wt% CO₂. Beyond that, the coal injection system and hydrogen chloride scrubber pump were considered in the

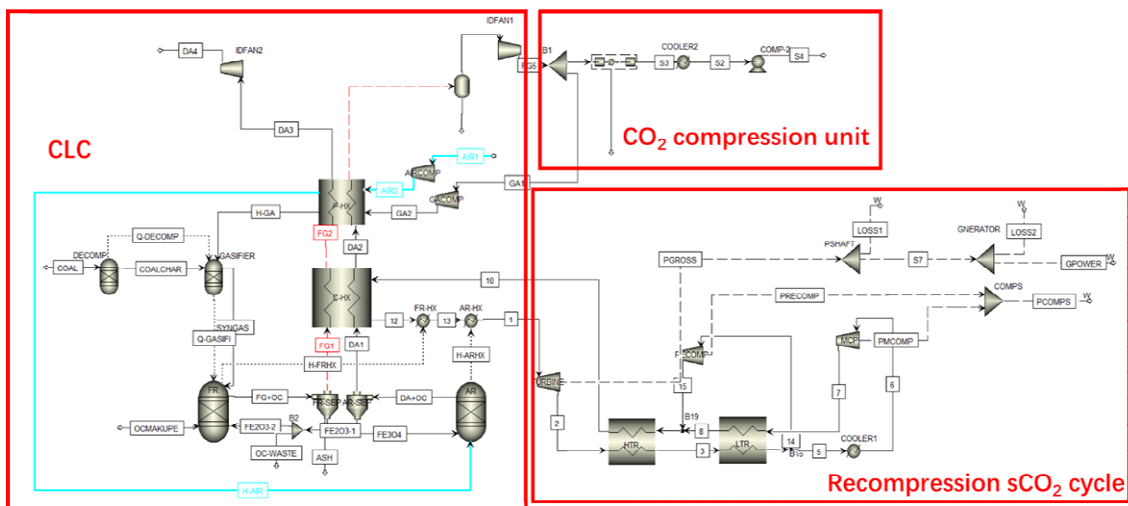


Figure 5-2 The process flow diagram of the proposed oxy-fuel combustion system with recuperated sCO₂ power cycle (No system is considered a black box.)

The design basis and assumptions of the reference CLC power plant described in Section 5.1.1 are utilised for the TEA calculation for the improved power plants, to ensure fair and valid comparisons.

At present, it is difficult to estimate the char conversion efficiency in the full-scale CDCLC plant [212]. 96% of char conversion efficiency is given in the reference plant associated with Fe₂O₃ as the OC. In Table 5-3, the char conversion efficiency of other OCs are estimated based on their characteristic [212], such as the manganese ore has better char gasification efficiency than the Fe₂O₃, so 97% of char conversion efficiency was assumed.

After the above consideration, the proposed CDCLC process was comprehensively simulated by Aspen Plus™ (Aspen Technology, Inc., Burlington, MA). The thermodynamic properties were estimated using Lee–Kesler–Plöcker property method (LK-PLOCK) for gas-processing in the sCO₂ power cycle, and the Peng–Robinson property method (Peng–Robinson cubic equation of state) was used to model all other process units. Pure component properties were obtained from the ASPENPCD, COMBUST, INORGANIC, SOLIDS, AQUEOUS, and PURE26 databanks in Aspen Plus™, and the General coal enthalpy model (HCOALGEN)

and Institute of Gas Technology coal density model (DCOALIGT) property models were used to achieve enthalpy and density, respectively, of coal and ash. The fuel reactor and air reactor were modelled by RGIBBS blocks based on the Gibbs free energy minimization. It is worth noting that, if the CLC Aspen model is a loop, a mole balance should keep the same between OC makeup rate fed into FR and OC waste discharged with fuel ash purge stream from AR.

5.2 Considerations for sensitivity analysis

From the above description and assumptions, the consideration for TEA of considered cases shows in this section. Section 5.3 and Section 5.4 show the results of operation and cost under the same techno-economic assumptions with the reference case. Section 5.5 presents several sensitivity analyses to study how the potential impact factors influence techno-economic performance indicators. In Figure 5-3, the relationship between the model assumptions or design specifications and performance indicators in the sensitivity analysis

The following potential impact factors were considered with their reasonable range.

- Turbine inlet temperature (TIT): between 550°C to 800°C with 50°C difference [202];
- Compressors outlet pressure (COP): between 150 bar to 300 bar [202];
- Char conversion efficiency in the fuel reactor: between 85% to 100% [178];
- OC attrition rate: its variation shows in Table 5-3 with directly impacting on OC makeup rate;
- Carbon tax: between 0 €/tCO₂ to 150 €/tCO₂;
- Discount rate: between 1.0% and 15.0%;
- CO₂ transportation, storage and monitoring unit cost: between 0 €/tCO₂ to 15 €/tCO₂;
- Capacity factor: between 85% to 100%;
- Fuel unit price: between 0 €/GJ to 6 €/GJ both coal and WP [213]; and

- OC unit cost: the range of each OC unit cost shows in Table 5-3, if not, it was estimated with $\pm 50\%$ fluctuation.

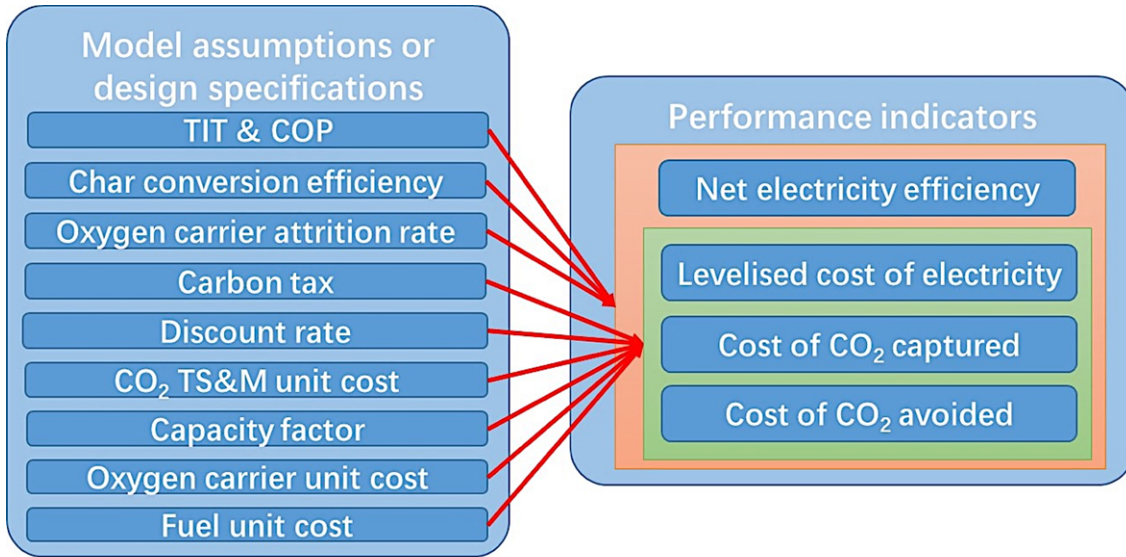


Figure 5-3 The relationship between the model assumptions or design specifications and performance indicators in the sensitivity analysis

5.3 Thermodynamic assessment

Table C-1 presents the technical performance of the CLC power plant with considered OCs, operating in the power cycle condition of 593°C and 240 bar for a fair comparison with the reference CDCLC power plant. Also, it was assumed the same fuel chemical energy input with 1566 MW_{th} in all considered cases. The OC makeup flow rate is estimated based on the OC attrition rate assumed in Table 5-3. Each component of the auxiliary electricity requirement of the CLC-sCO₂ power plant is estimated based on the reference CDCLC power plant.

Regarding the net electricity efficiency, comparing with a similar size CDCLC steam Rankine cycle power plant [214], most of the considered coal-fuelled CLC performed better than the improved process introduced in Section 5.1.2, mainly attributed to employing the recompression sCO₂ power cycle. Only in the cases of coal-fuelled CLC with CaSO₄ and Fe₂TiO₃ as the OC, there are 0.57%_{HHV} point and

0.12%_{HHV} point net efficiency penalty, respectively, owing to the endothermic decomposition reaction of limestone and ilmenite, respectively, as the makeup injected into the fuel reactor.

The auxiliary electricity requirements of CLC-sCO₂ power plants in all considered OCs generally occupy just over 14% in coal-fuelled cases and just over 17% in WP-fuelled cases, respectively, of gross electricity generation. Of these, the power consumption of CO₂ compressor trains accounts for over 50% of total auxiliary electricity requirements for achieving the required pressure of CO₂ transport and storage. However, in the literature, regarding the conventional supercritical PC cases without employing the CO₂ capture system [215], the auxiliary electricity requirements account for only 5.2% of its gross electricity generation. This mainly attributes to without considering CO₂ compressors for CO₂ transport and storage, and slightly attributes to without considering the OC handling system (0.1-0.8% of gross electricity generation) and enhancer gas recycle compressors (1.1-1.2% of gross electricity generation). Furthermore, if considering CCS in the conventional supercritical PC case [215], around 17% of gross electricity generation is consumed by the auxiliary equipment mainly owing to large power consumptions of the additional Fluor Econamine FG PlusSM process for CO₂ removal and CO₂ compressors for CO₂ transport and storage.

Except for the above analysed coal-fuelled cases, the biomass-fuelled cases have around 3.5%_{HHV} net efficiency penalty owing to the fuel characteristic of biomass, which also reveals on them lower boiler heat-exchange efficiency, the higher specific fuel chemical energy consumption, and specific fuel consumption. Furthermore, the CO₂ product purity and the CO₂ capture efficiency in all considered cases meet the design requirements with over 95mol% and over 90%, respectively. Around 3.5 million tons of CO₂ is captured per year in coal-fuelled cases, and the biomass-fuelled cases are higher than that because of the higher fixed char content in the biomass, which also reflects in the value of specific CO₂ captured. In addition, biomass-fuelled cases achieved negative specific CO₂

emission lower than $-1000 \text{ gCO}_2/\text{kWh}_{\text{el, net}}$ by virtue to the concept of BECCS with a potential benefit of an additional revenue stream from the ETS.

In terms of the thermodynamic performance of OCs considered in CLC with sCO_2 power cycle, it obviously reveals the differences between the natural OCs and the manufactured OCs. For instance, in Figure 5-4, most of the net electricity efficiency of the manufactured OCs cases have better value than the natural OCs cases owing to the previously mentioned reason that the endothermic decomposition reaction of OC makeup exists in the fuel reactor. Correspondingly, coal consumption per net electricity production in the manufactured OCs cases keep the same, rather than that in the natural OCs cases increases 2.9% and 1.6% of limestone and ilmenite, respectively, as the OC makeup. Regarding the auxiliary electricity requirement, the value of each case is around 6%. Another key thing to notice is that the assumed char conversion efficiency has impacted numerous thermodynamic indicators, such as CO_2 product purity, CO_2 mass flow rate in CO_2 product, specific CO_2 capture and CO_2 capture efficiency are positively impacted, but CO_2 emission rate and specific CO_2 emission are negatively impacted.

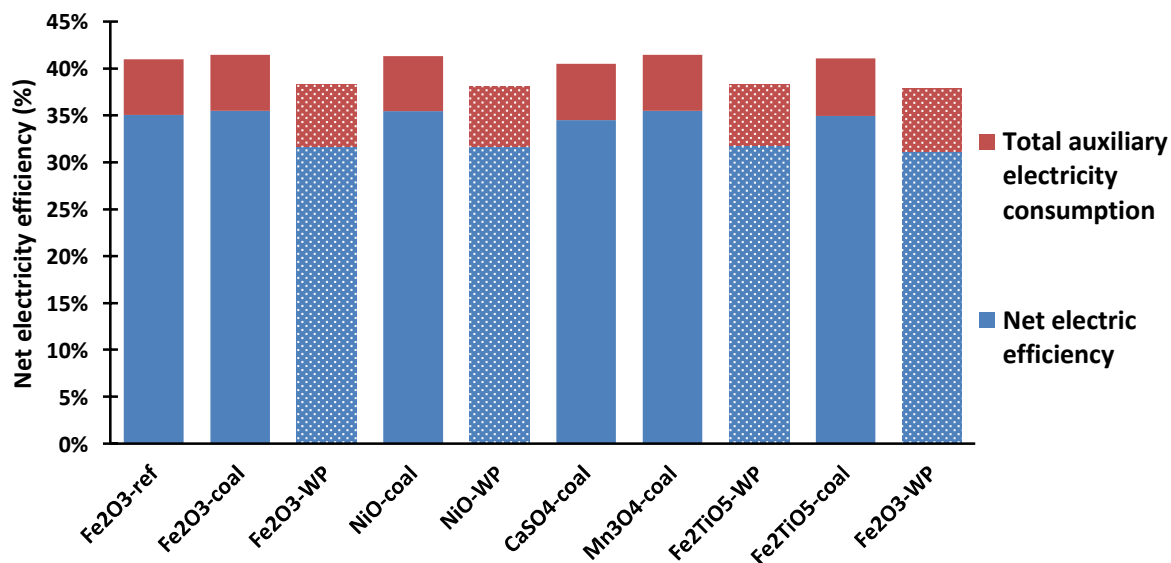


Figure 5-4 Net electricity efficiency and total auxiliary electricity consumption of considered cases in Chapter 5 (The fully colour-filled area represents the coal-

fuelled case, and the partially colour-filled area represents the biomass-fuelled case.)

5.4 Economic assessment

Table C-2 shows the details of economic assessment results of all considered cases, including the detailed distributions of all considered cases and the capital COE, fixed O&M COE, variable O&M COE, and fuel COE. Figure 5-5 shows the detailed costs of the sCO₂ power cycle system for each case. Figure 5-6 shows the LCOE distribution of the considered CLC cases. Figure 5-7 shows AC and CC of considered CLC cases.

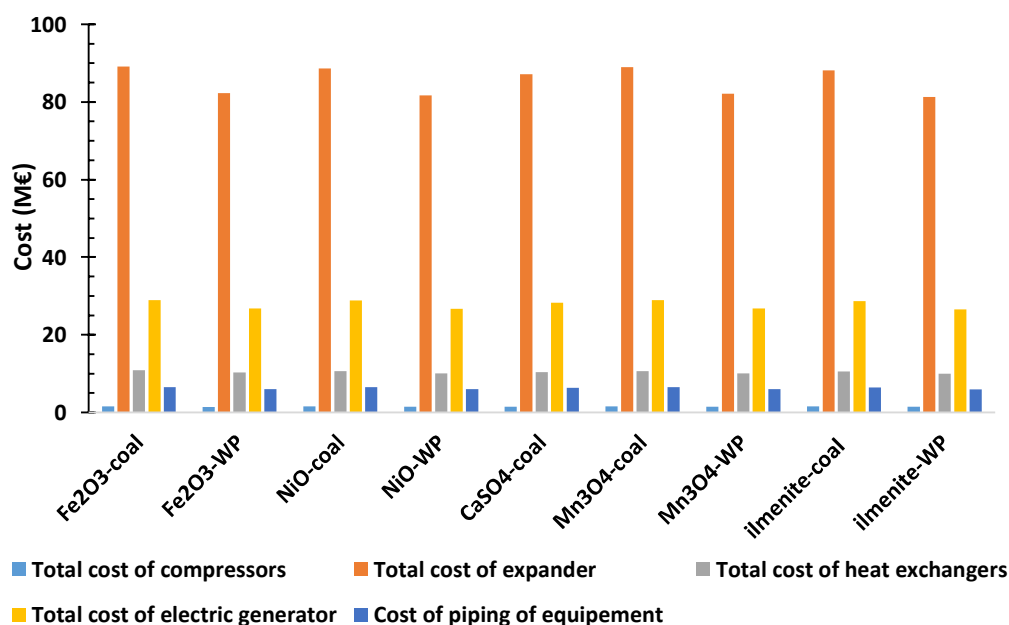


Figure 5-5 Cost distributions of sCO₂ power cycle system

Regarding the cases considering the manufactured OCs, the specific capital cost and capital COE of the coal-fuelled CLC were improved by both 1.8% compared with the reference case. This proves that replacing the conventional steam Rankine cycle with a sCO₂ power cycle brings substantial economic benefits as the latter improves the net electricity efficiency and reduces the capital cost of the entire system. However, in the natural OC based CLC cases, their specific capital

cost and capital COE increase by 5% to 8% which is consistent with a natural OC-based CLC power plant [178]. Furthermore, if the WP is fuelled in the considered CLC-sCO₂ power plants with manufactured and natural OCs, these figures increase between 16% and 19%, which owing to the WP have higher moisture and furthering lower net electricity efficiency. Regarding the detailed cost distributions of the sCO₂ power cycle system, it reveals that the most expenditure costs on the expander with about 65% in the total cost of sCO₂ power cycle system in considered coal- and biomass-fuelled cases.

In addition, the fixed O&M COE does not have a significant difference in all cases which is stable around 11 €/MW_{el}h. What is more, in the coal-fuelled case of the Manganese ore as the OC makeup, its variable O&M COE with 13.9 €/MW_{el}h is the lowest in all considered coal-fuelled cases, which is notably half of the coal-fuelled cases with manufactured OCs as the makeup. This results from the common impact of a relatively lower attrition rate and sizeable lower delivered price of the Manganese ore. For example, the attrition rate of manufactured Fe₂O₃ is around 10 times lower than that of the Manganese ore, but its delivered price is at least 20 times, even 90 times, higher than that of the Manganese ore, which makes a significantly higher OC cost in variable O&M cost and furthering significantly higher LCOE in case of manufactured Fe₂O₃. Likewise, this effect is relatively small in the case of ilmenite as the OC makeup, which is expensive than the Manganese ore case, but still lower than the manufactured Fe₂O₃ case. However, if the biomass was applied in all cases, they were worse than themselves coal-fuelled cases. The variable O&M COE in investigated cases has a large difference with the details shown in Table C-2, which owing to the OC COE primarily attributed to the attrition rate and OC price.

Another key thing to remember, 0 €/tCO₂ of the carbon tax is initially used in the variable O&M cost, but it could be uncertain if it is introduced in the ETS. By virtue to the advantages of BECCS, the variable O&M cost in biomass-fuelled cases

could be substantially reduced under a certain carbon tax, furthering could reduce the LCOE.

The fuel COE has not shown the difference in considered coal-fuelled cases been steady at 25.2 €/MW_{el}h, but it is around 2 times in the biomass-fuelled cases than in coal-fuelled cases primarily, attributing to their lower η_{net} and a higher fuel price per gigajoule of WP.

Finally, the LCOEs of the natural OC cases are, in general, better than that of the manufactured OC cases, based on the same fuel cases. From the above description, it is found that the LCOE is reduced in all coal-fuelled cases when compared to the reference case. The largest reduction of up to 2.1% was observed in the natural OC cases. The best case of LCOE is 96.8 €/MW_{el}h in the cases of Manganese ore as OC makeup with 12.4 €/MW_{el}h (11.4%) LCOE reduction. For the biomass-fuelled cases, the Manganese ore case achieves the best LCOE of 129.8 €/MW_{el}h, which is over 12.6 €/MW_{el}h lower than the best manufactured case (Fe₂O₃). However, the biomass-fuelled Mn₃O₄ case showed significantly higher LCOE due to the 3.8%_{HHV} of net efficiency penalty compared with the coal-fuelled Mn₃O₄ case. The 3.8% is calculated based on the data in Table C-2. It equals to the difference between 35.49% and 31.75%, which because of the lower heating value and higher moisture of biomass.

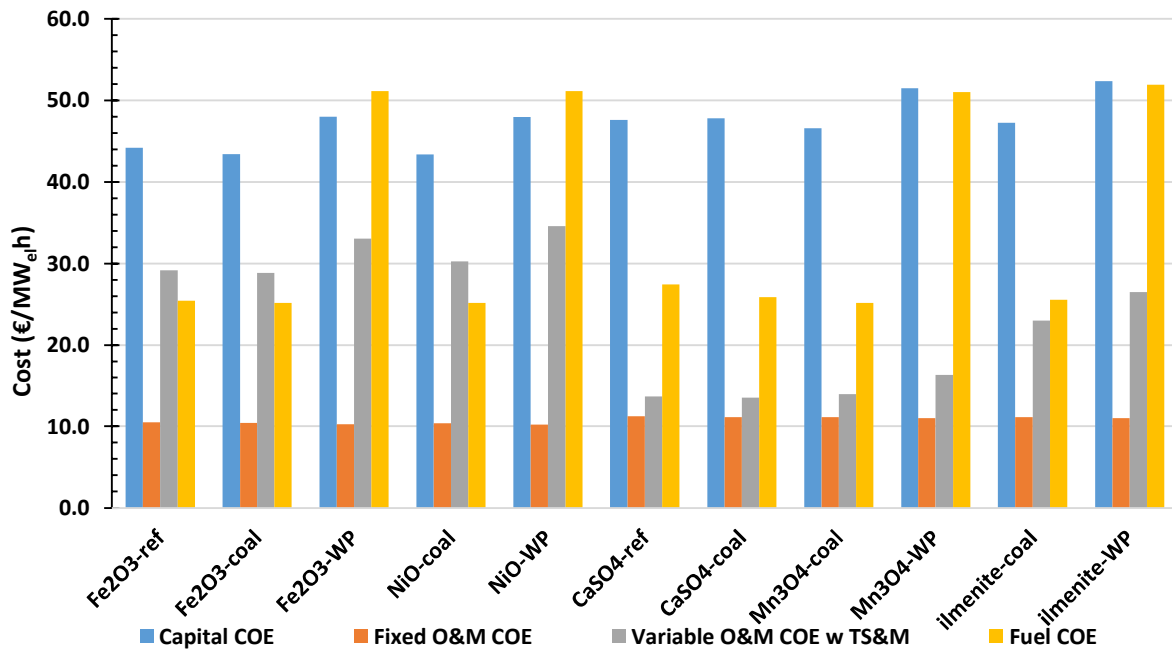


Figure 5-6 LCOE distribution of the considered CLC cases

At the same time, comparing with the reference CLC case with 90.9 €/tCO₂ of AC and 74.9 €/tCO₂ of CC, the AC and CC are reduced by 1.7 €/tCO₂ and 0.68 €/tCO₂, respectively, by just replacing the steam Rankine cycle to sCO₂ power cycle. Based on that, if continuously replacing the manufactured OC of Fe₂O₃ with the manufactured OC of NiO, the AC and CC are increased by 3.6 €/tCO₂ and 4.4 €/tCO₂, respectively, owing to its higher LCOE, higher CO₂ emission rate and lower CO₂ capture rate. However, the AC and CC of the limestone, manganese ore and ilmenite reduced to 76.9 €/tCO₂ and 61.8 €/tCO₂, 74.0 €/tCO₂ and 60.4 €/tCO₂, and 88.1 €/tCO₂ and 66.6 €/tCO₂, respectively. Likewise, if replacing to the fuel of biomass in all considered cases, their AC achieves sizeable reduction with lower to 48.3 €/tCO₂, which mainly because of the significantly negative CO₂ emission rate offsets the higher LCOE. Notably, the CCs of biomass-fuelled cases are expensive than the reference case mainly owing to the higher LCOE and higher specific CO₂ capture of the reference case. Nonetheless, introducing the ETS with the carbon tax is expected to reduce the CC in biomass-fuelled cases, which shows in the following sensitivity analysis.

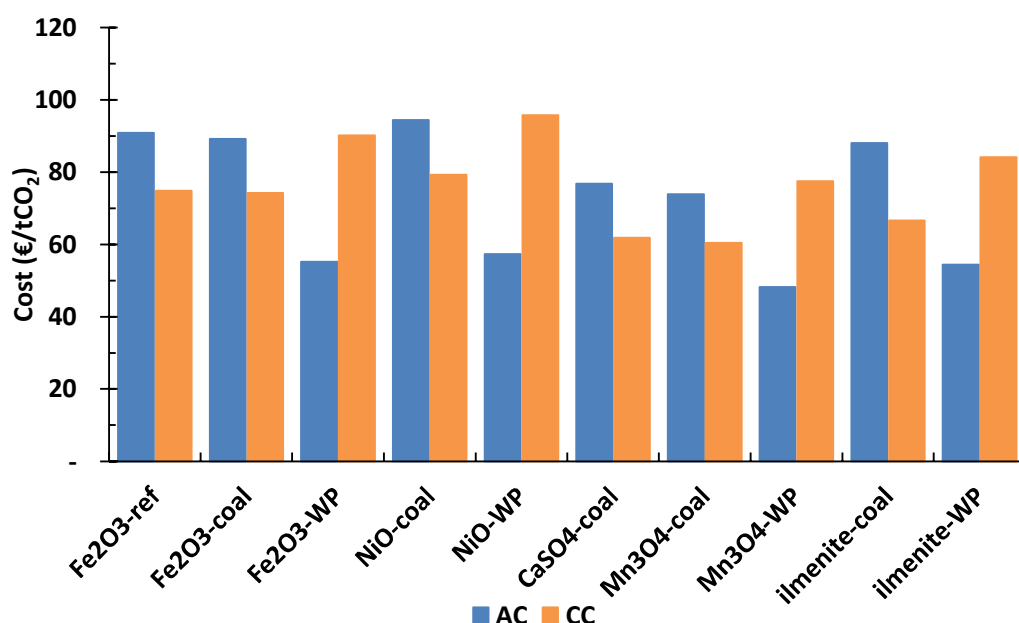


Figure 5-7 AC and CC of considered CLC cases

5.5 Sensitivity assessment

From the assessments in Section 5.3 and 5.4, it was found that manganese ore as the OC performed well among other OCs due to the tiny reduction of net electricity efficiency and sizeable reduction of LCOE. Therefore, it was taken as the representative comparing with the reference CDCLC power plant using manufactured Fe_2O_3 as the OC. Meanwhile, other OCs is analysed in the attached supplementary document. The sensitivity analysis is divided into two sections. The first section focuses on the technical aspect (Section 5.5.1) through adjusting four technical impact factors in Aspen Plus™, including TIT, COP, char conversion efficiency, and OC efficiency. Another section focuses on the economic aspect (Section 5.5.2). During the economic calculation, six economic-related impact factors were found directly impacting the economic performance of CLC, including carbon tax, discount rate, CO_2 transport, storage and monitoring unit cost, capacity factor, OC unit cost, and fuel unit cost.

5.5.1 Technical performance factors

Increasing the TIT & COP is generally recognized as a way of improving the efficiency of the power cycle [145]. To analyse the coal-fuelled case first, in Figure 5-8 a), it was found that the net electricity efficiency was increased by increasing the TIT with the gradual reduction in effectiveness. When the TIT is above 600°C with investigated COP, this net electricity efficiency is surpassed that of the reference CDCLC power plant. However, raising the COP did not achieve the expected outcomes. It was shown that there is an optimal net electricity efficiency when the COP is around 200 bars by keeping the same temperature. And if the TIT increases to 750°C and 800°C, respectively, the optimal net electricity efficiency is by increasing the COP to 250 bars. From the above, by keeping the same TIT, it could be conjectured that the optimal net electricity efficiency gradually moves from the 200 bars to the 250 bars. Moreover, in Figure 5-8 b) c) d), the LCOE, AC and CC were influenced by net electricity efficiency, and were completely superior to the reference CLC power plant in all considered cases. Meanwhile, it was found that, apart from the CC, the LCOE and AC were obviously affected by the net electricity efficiency with the optimal results in the COP of the 200 bars or the 250 bars.

On the subsequent analysis of the biomass-fuelled cases (Figure 5-9), it reveals that the curve variation tendency (not referring to the number variation) of the net electricity efficiency, LCOE, AC and CC in coal-fuelled cases is similar to those in biomass-fuelled cases. In Figure 5-9 a), the net electricity efficiency of the biomass-fuelled cases in most of TIT & COP cases are below that of the reference CDCLC power plant, due to the characteristic of biomass resulting in the lower boiler efficiency. The lower net electricity efficiency directly influences the LCOE resulting in all values above the reference LCOE, shown in Figure 5-9 b). This further impacts the CC with higher outcomes in Figure 5-9 d) comparing with the coal-fuelled cases in Figure 5-8 d). Nevertheless, it achieved remarkable AC in Figure 5-9 c) that is significantly lower than the reference case, resulting from the

inherent decarbonization ability by using biomass as fuel. From the above investigations, the overall performance of biomass-fuelled CLC is worse than that of coal-fuelled CLC. The further studies were conducted for finding a potential way to enhance the competitiveness of the biomass-fuelled CLC power plant.

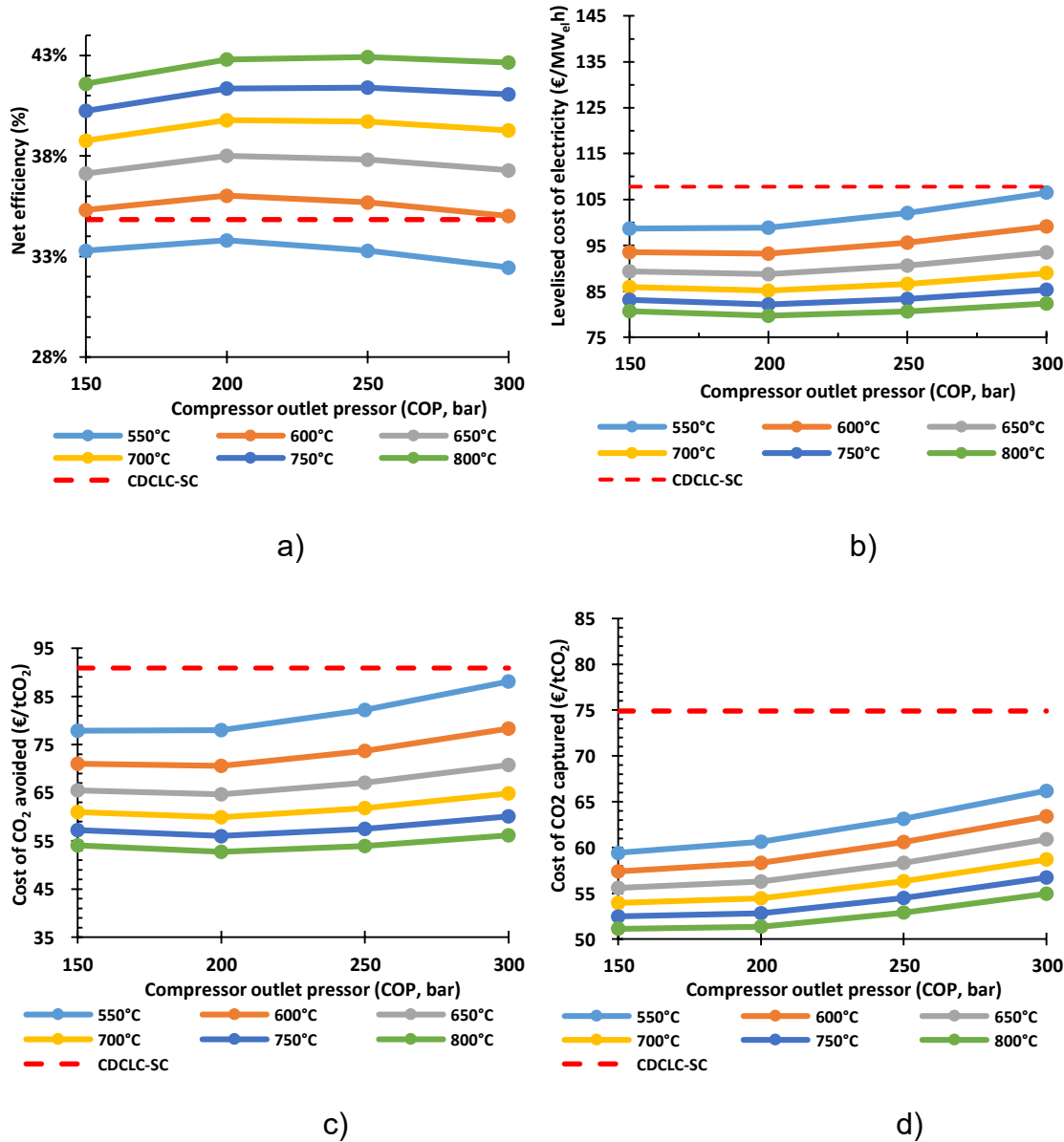


Figure 5-8 Influence of turbine inlet temperature and compressor outlet pressure on a) net electricity efficiency, b) levelised cost of electricity, c) cost of CO₂ avoided,

and d) cost of CO₂ captured in the coal-fuelled CLC power plant with Mn ore as the oxygen carrier

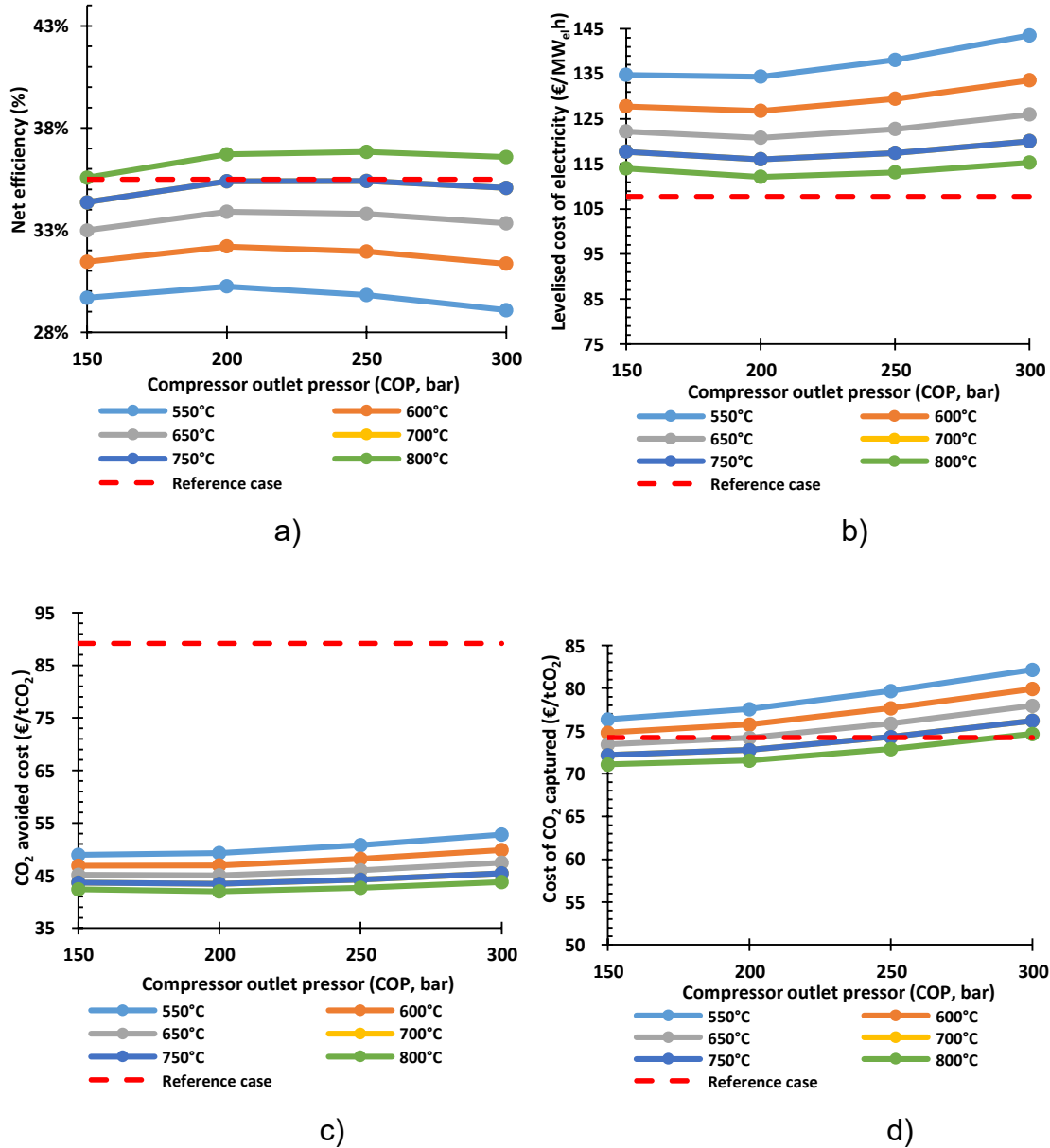


Figure 5-9 Influence of turbine inlet temperature and compressor outlet pressure on a) net electricity efficiency, b) levelised cost of electricity, c) cost of CO₂ avoided, and d) cost of CO₂ captured in the biomass-fuelled CLC power plant with Mn ore as the oxygen carrier

In Figure 5-10, an interesting result was found after the sensitivity study on the char conversion efficiency in the coal-fuelled case was performed. Increasing the char conversion efficiency from 90% to 100% intuitively results in a net efficiency penalty of 0.3% owing to the power consumption increase of the CO₂ compressor trains, which directly results in a 1.4% increase of LCOE. The fundamental reason is that less CO₂ passes the CO₂ compressor trains resulting from the unconverted carbon is transferred into the air reactor and is burned with CO₂ emitted. Although the relatively negative variation tendency on net electricity efficiency and LCOE was described above, their values in investigated cases of char conversion efficiency all performed well than the reference CDCLC power plant. Simultaneously, the emitted CO₂ in depleted air from the air reactor negatively impacts the CO₂ capture efficiency of the entire system. Moreover, through varying the char conversion efficiency from 90% to 100%, positive results were found that a 9.5% reduction of AC and 10.6% reduction of CC by virtue to that the influence from complete char conversion in the fuel reactor is greater than that from the net efficiency penalty. Similarly, the values of AC and CC are all below the reference case with the difference on 18.9 €/tCO₂ of AC and 16.2 €/tCO₂ of CC, respectively.

The similar variation tendency was found in the biomass-fuelled CLC power plant with a 0.35% reduction of the net electricity efficiency, a 1.6% increase of LCOE, a 4.4% reduction of AC and a 10.7% reduction of CC by adjusting the char conversion rate from 90% to 100%. Comparing with the reference case, the poor performance of the net electricity efficiency, LCOE and CC reveals in Figure 5-10 a) and b). Surprisingly, in Figure 5-10 b), the values of AC are largely lower than the reference case with around a 35% reduction of AC by virtue to the concept of BECCS.

The OC attrition rate is treated as a key performance factor and has received more attention in the study of CLC, which is expected to influence the techno-economic performance. True to the expected, through the modelling and calculation, the OC attrition rate directly decides the OC makeup rate. It further impacts the power

consumption on OC particle handling and OC total cost, which finally results in the variation of the four key performance indicators shown in Figure 5-11. It was found that, with increasing of OC attrition rate from 0 to 0.5%, the variations are 0.55% points of net electricity efficiency penalty, 9.2 €/MW_{el}h increase of LCOE, 12.0 €/tCO₂ increase of AC and 9.8 €/tCO₂ increase of CC. Although their variation tendency gets worse, they approach to the reference case level. It indicates that the OC attrition rate is essential and non-ignorable to impact the overall performance of the CLC power plant.

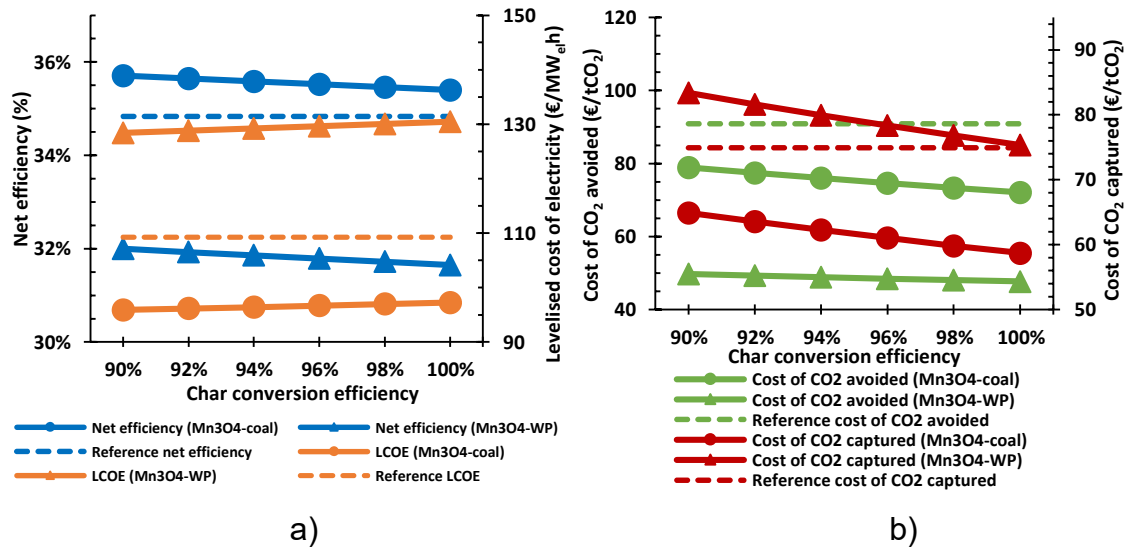


Figure 5-10 Influence of char conversion efficiency on a) net electricity efficiency and levelised cost of electricity, and b) cost of CO₂ avoided and cost of CO₂ captured in the CLC power plant with Mn ore as the oxygen carrier

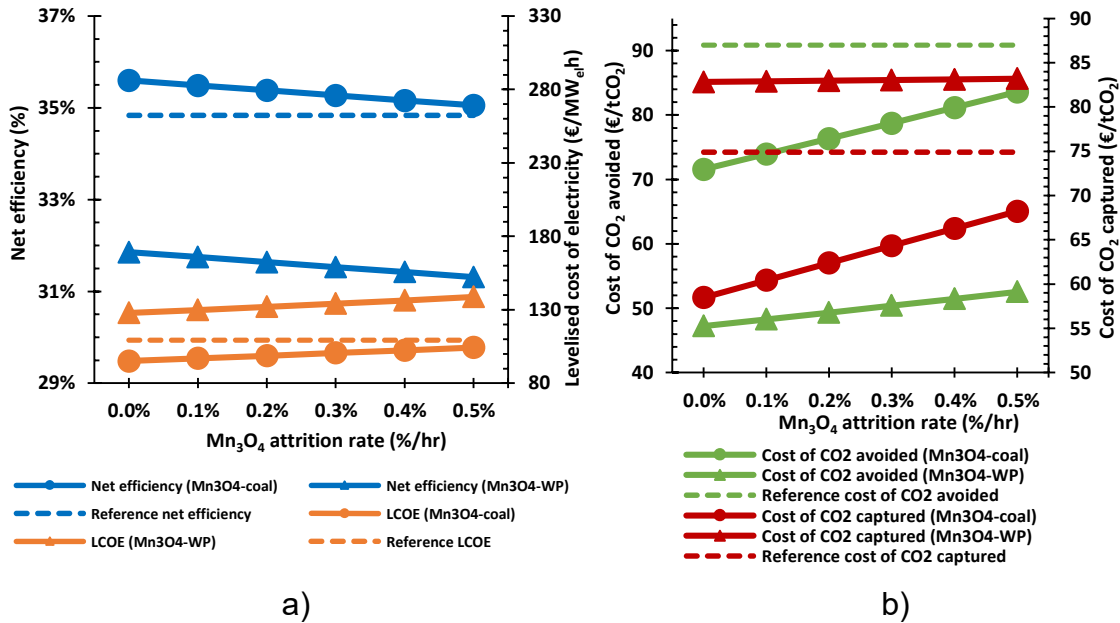


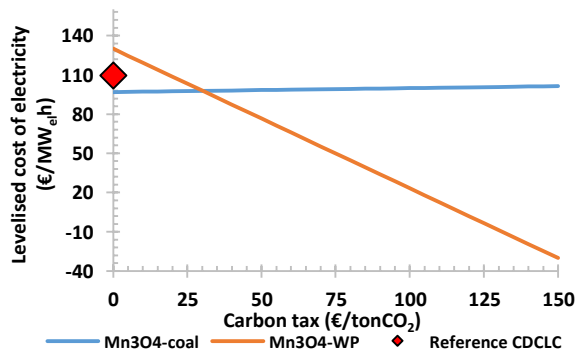
Figure 5-11 Influence of oxygen carrier attrition rate on a) net electricity efficiency and levelised cost of electricity, and b) cost of CO₂ avoided and cost of CO₂ captured in the CLC power plant with Mn ore as the oxygen carrier

In the biomass-fuelled cases, in Figure 5-11 their overall performance on net electricity efficiency, LCOE and CC are worse than the coal-fuelled cases owing to the characteristic of biomass. Since biomass as the fuel, its excellent performance on AC is by virtue to the negative emission of BECCS.

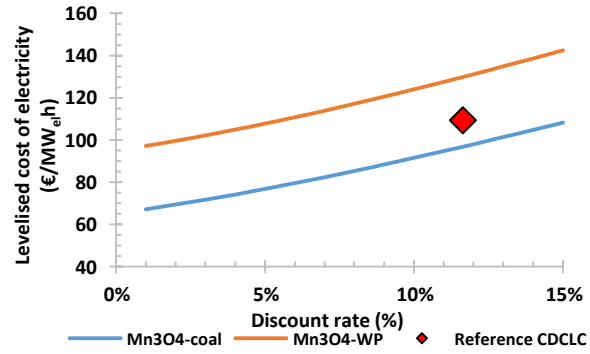
5.5.2 Economic performance factors

In Figure 5-12, Figure 5-13 and Figure 5-14, the LCOE, AC and CC were investigated by the six economic impact factors. It demonstrated that if Manganese ore and sCO₂ power cycle are applied, the LCOE, AC and CC in both coal- and biomass-fuelled CLC power plants are positively correlated by discount rate, OC price and fuel price. However, they are negatively correlated by the capacity factor (Figure 5-12 d), Figure 5-13 d) and Figure 5-14 d)). In Figure 5-12 a), it is worth noting that the carbon tax has specificity on the LCOE of the biomass-fuelled. On an increase of the carbon tax, the LCOE of the coal-fuelled was increased, but the LCOE of the biomass-fuelled is more sensitive with decrease. It decreased

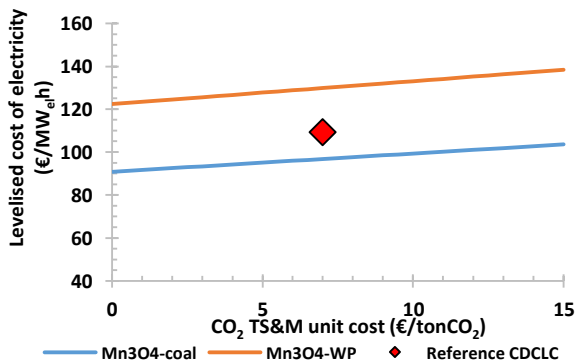
dramatically even below 0 €/MW_{el}h which means the biomass-fuelled CLC power plant cost 0 € to generate per kilowatt-hour electricity when the carbon tax is 121.8 €/tCO₂. A significant point intersection shown in Figure 5-12 a) represents the biomass-fuelled CLC power plant become feasible at carbon tax over 30.1 €/tCO₂. In Figure 5-13 a), the AC of the biomass-fuelled has negatively correlated by the carbon tax, rather than the coal-fuelled. Moreover, it is more sensitive and greatly lower than coal-fuelled. In general, the AC of the biomass-fuelled CLC is better than the coal-fuelled one. In Figure 5-14 a), the point intersection represents the CC of biomass-fuelled CLC is better than the coal-fuelled if the carbon tax is above 16.6 €/tCO₂. Regarding the CO₂ TS&M unit cost, it positively correlated with the LCOE and AC, but does not affect the CC because the CO₂ TS&M unit cost has not been calculated in the CC.



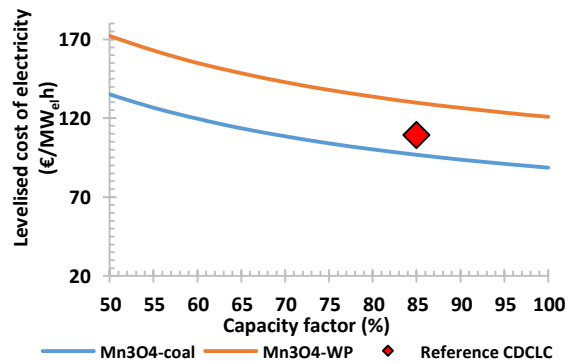
a)



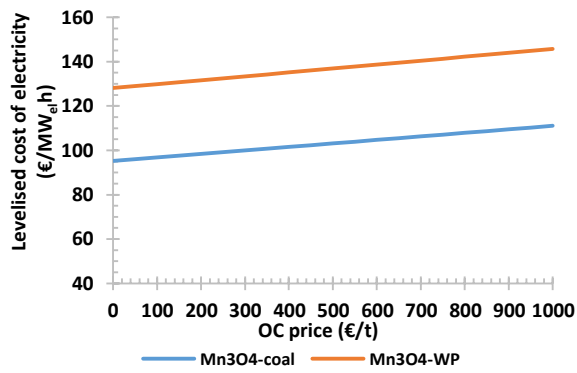
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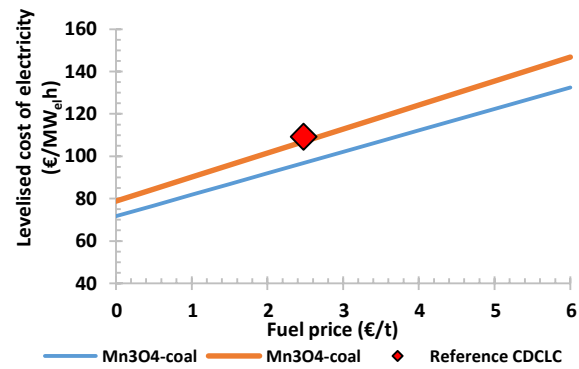
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d)

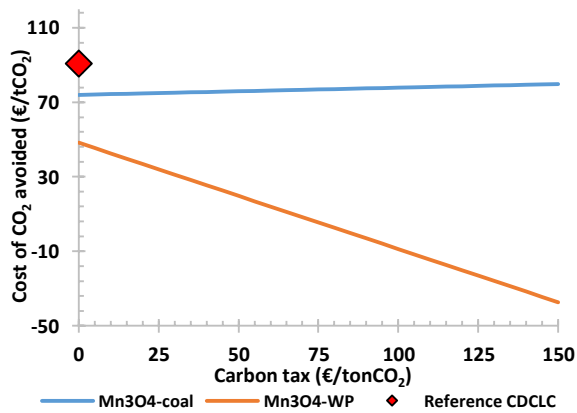


e)

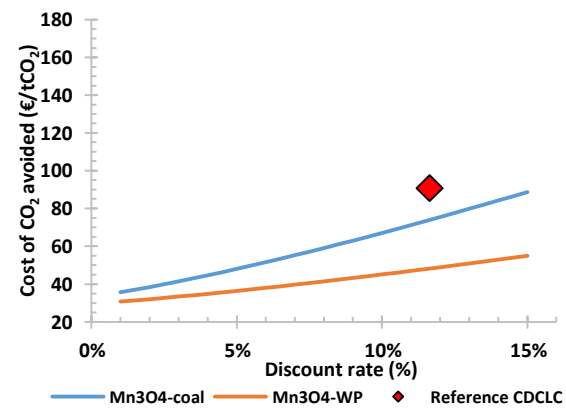


f)

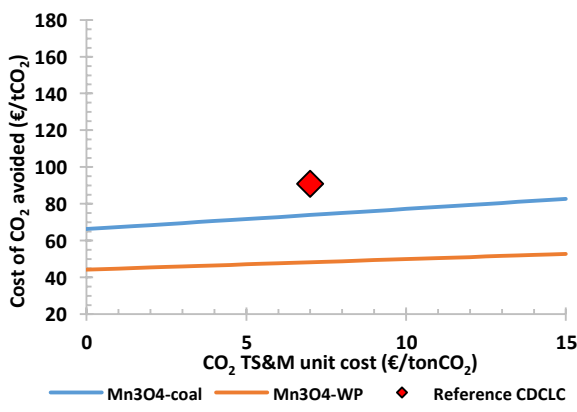
Figure 5-12 Influence of a) carbon tax, b) discount rate, c) CO₂ transport, storage and monitoring unit cost, d) capacity factor, e) oxygen carrier unit cost, and f) fuel unit cost on levelised cost of electricity of CLC power plant with Mn ore as the oxygen carrier



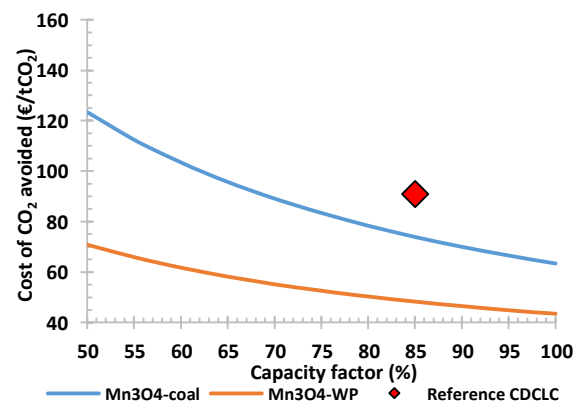
a)



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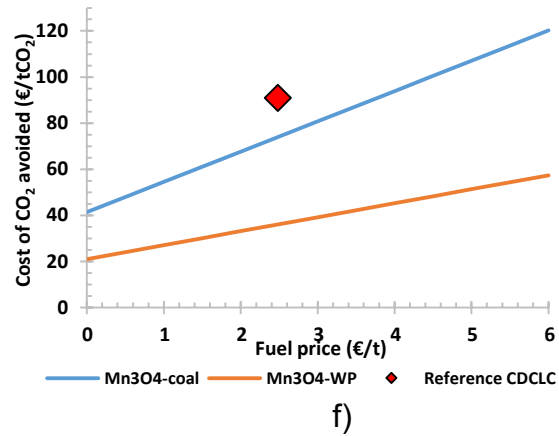
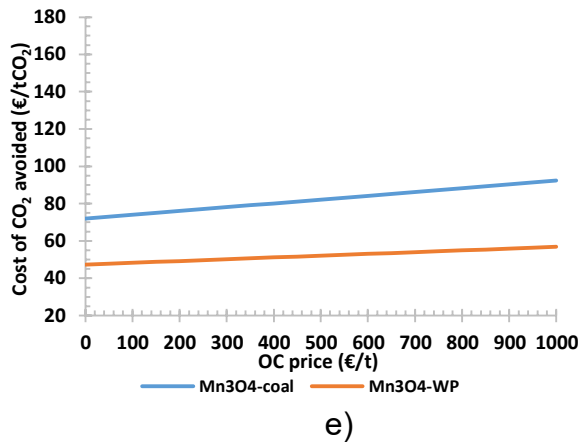
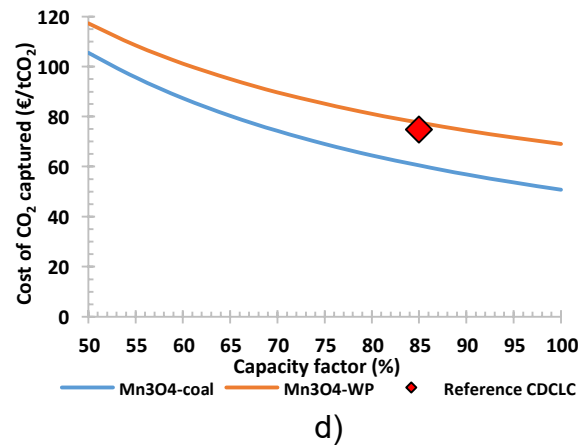
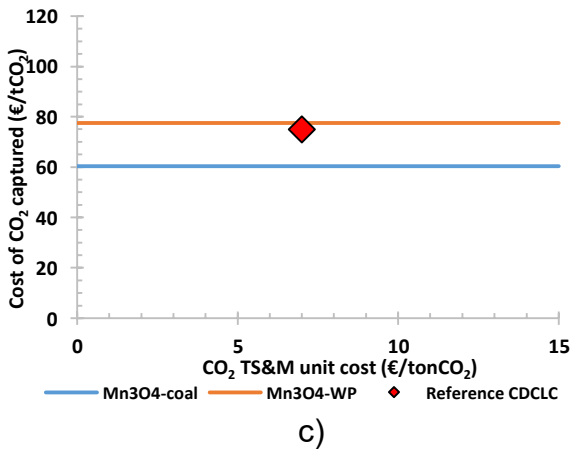
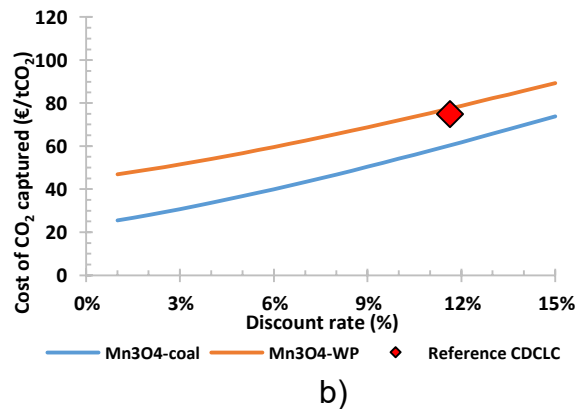
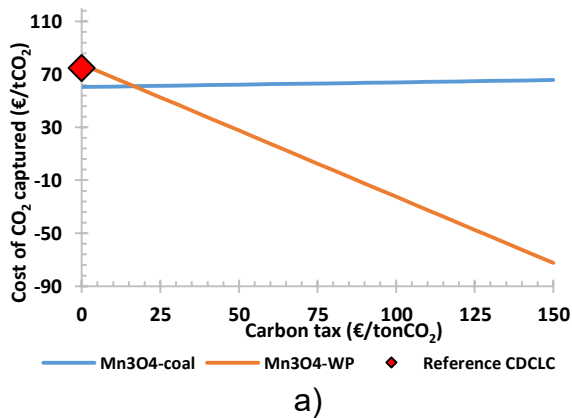


Figure 5-13 Influence of a) carbon tax, b) discount rate, c) CO₂ transport, storage and monitoring unit cost, d) capacity factor, e) oxygen carrier unit cost, and f) fuel unit cost on cost of CO₂ avoided of the CLC power plant with Mn ore as the oxygen carrier



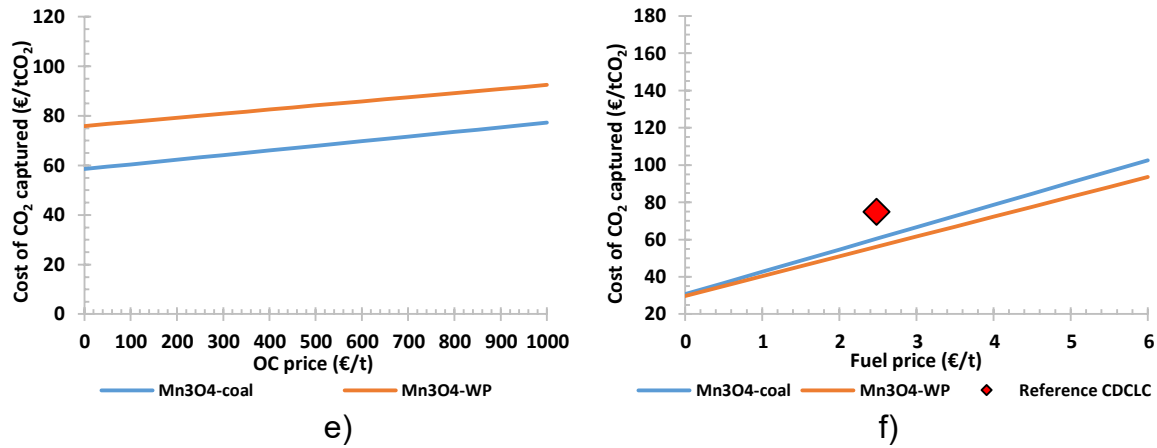


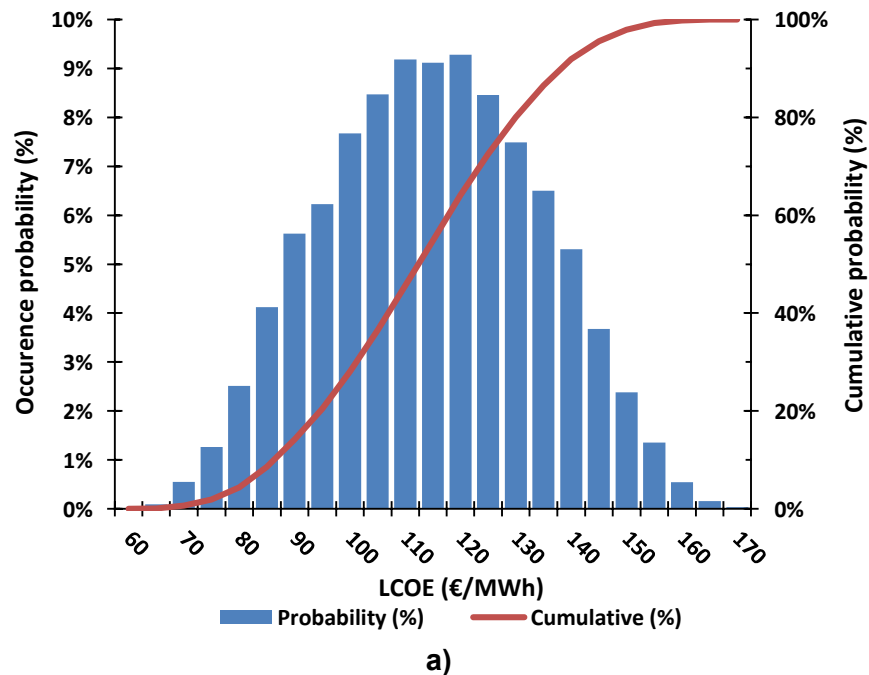
Figure 5-14 Influence of a) carbon tax, b) discount rate, c) CO₂ transport, storage and monitoring unit cost, d) capacity factor, e) oxygen carrier unit cost, and f) fuel unit cost on cost of CO₂ captured of the CLC power plant with Mn ore as the oxygen carrier

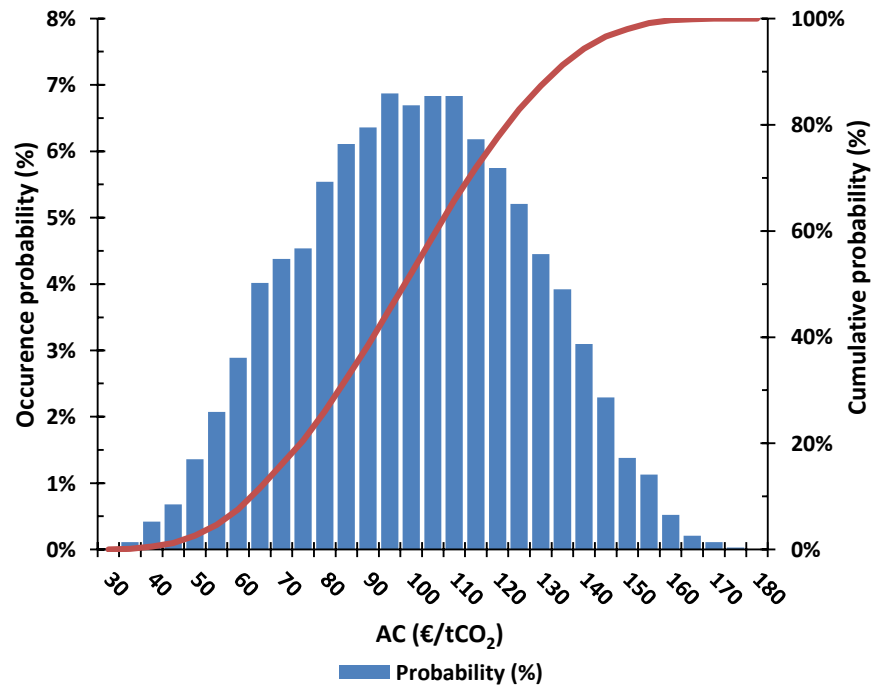
Through the above, the carbon tax is a critical factor associated with the European Union emissions trading system (EU ETS), which expects to impact biomass-fuelled CLC more competitively than the coal-fuelled. The current CO₂ European emission allowance is 25.15 €/MW_{el}h [204]. Additionally, the carbon taxes (updated April 2020) in the world have considerable differences, such as 106 €/MW_{el}h in Sweden, 44 €/MW_{el}h in France, 25 €/MW_{el}h in Canada, 20 €/MW_{el}h in the United Kingdom, 12 €/MW_{el}h in New Zealand, which decided the application feasibility of biomass-fuelled CLC power plant in the corresponding countries.

6 Probability economic assessment

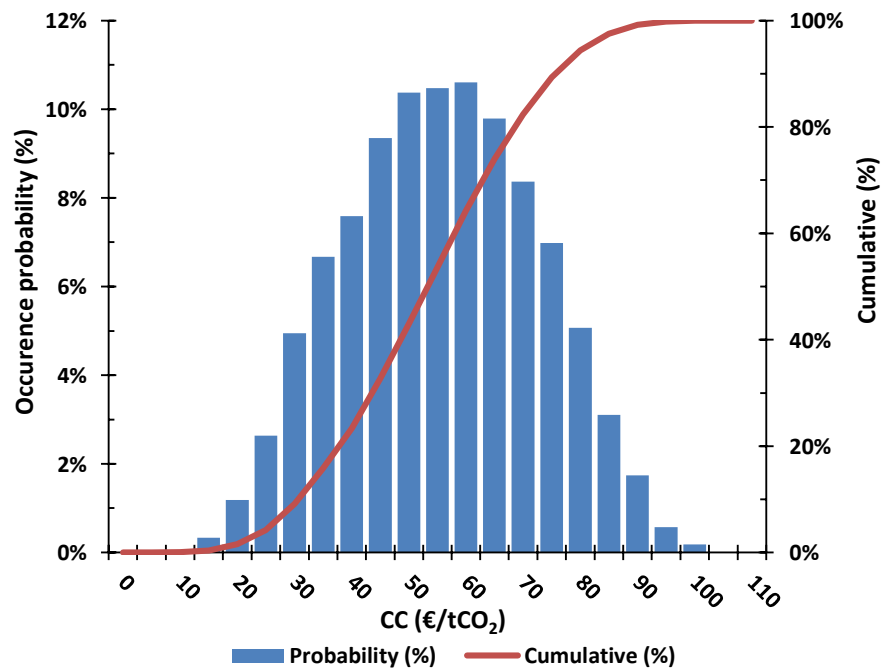
This chapter presents the probability of economic performance based on the two proposed systems studied in Chapter 4 and Chapter 5. Since the previous works have different assumptions for TEA which is not fair for comparison, assumptions, such as capacity factor and lifetime, are unified in their economic model for this chapter. Meanwhile, the same operation condition of the power cycle is assumed to be 593°C and 240 bar. These unified parameters reference a NETL report [102] for a fair comparison.

Figure 6-1 to Figure 6-4 show the occurrence and cumulative probability for the economic performance of considered oxy-fuel combustion and CLC power plants with the sCO₂ power cycle, including LCOE, AC and CC. The stochastic analysis has revealed that the statistical distribution of all these economic performance indicators is close to the normal distribution. Through analysing Figure 6-1 to Figure 6-4, the key probability indicators for economic performance on the considered cases, including minimum value, maximum value, mean value and standard deviation, are summarised in Table 6-2 and Table 6-3.



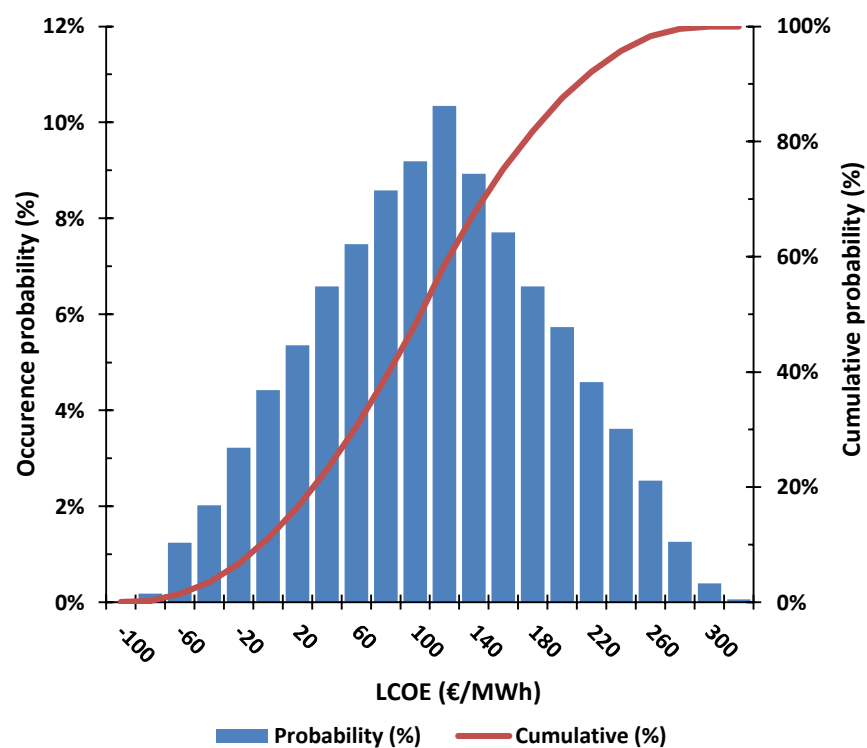


b)

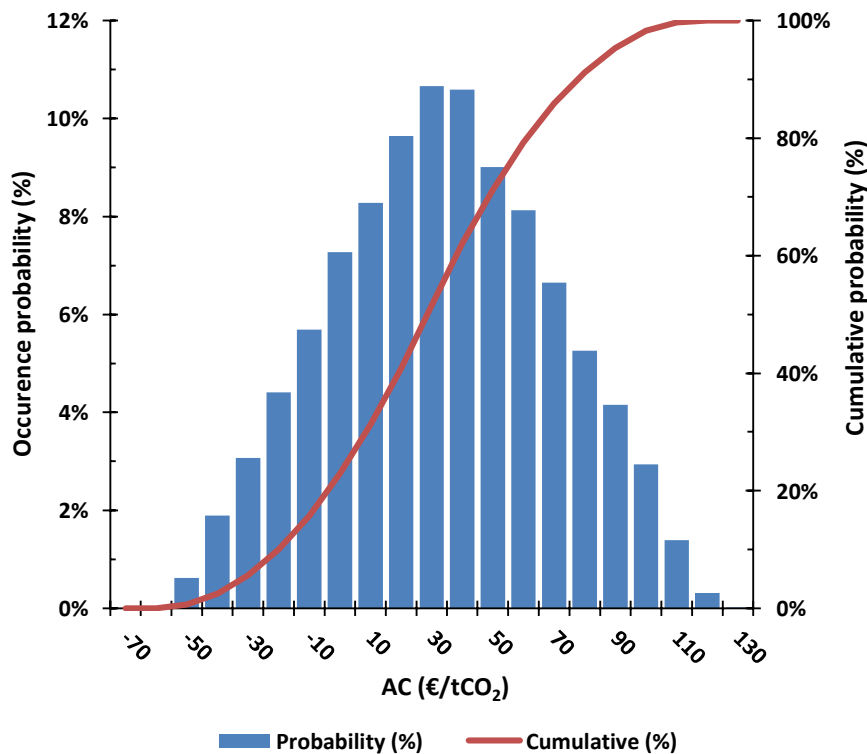


c)

Figure 6-1 Occurrence and cumulative probability for a) levelised cost of electricity, b) cost of CO₂ avoided, c) cost of CO₂ captured of the coal-fuelled oxy-fuel combustion power plant with the sCO₂ power cycle



a)



b)

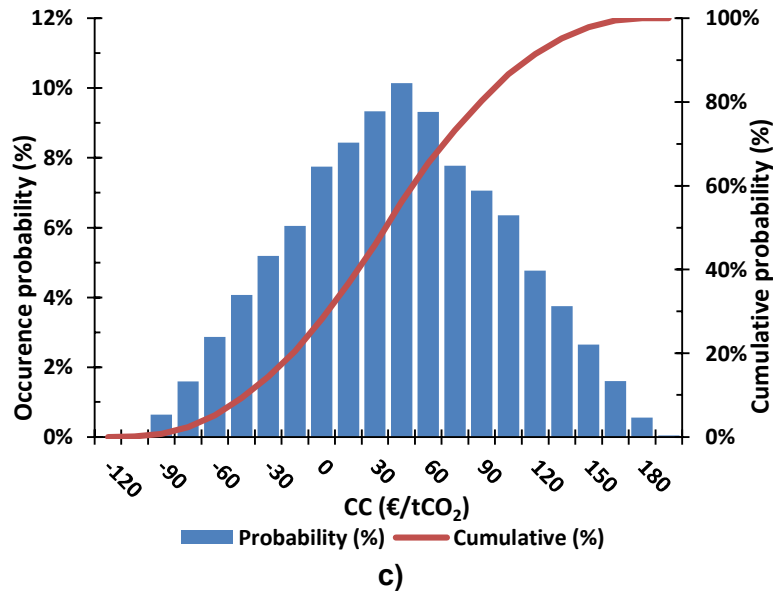
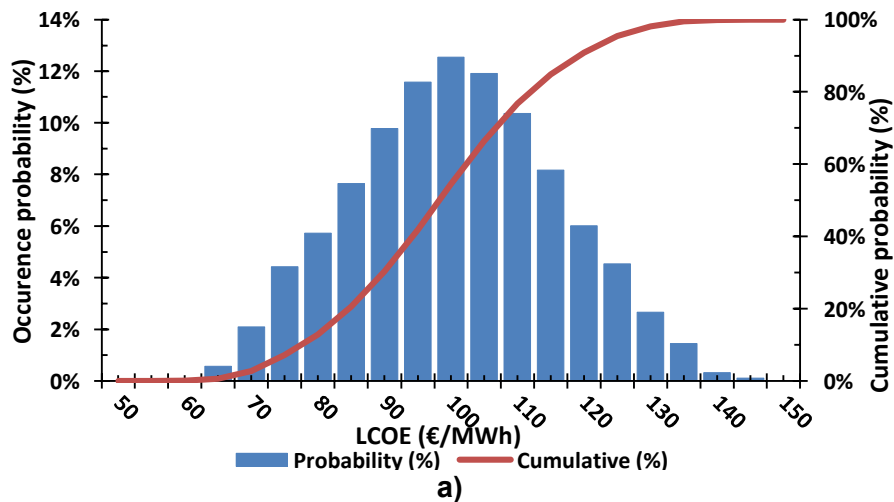
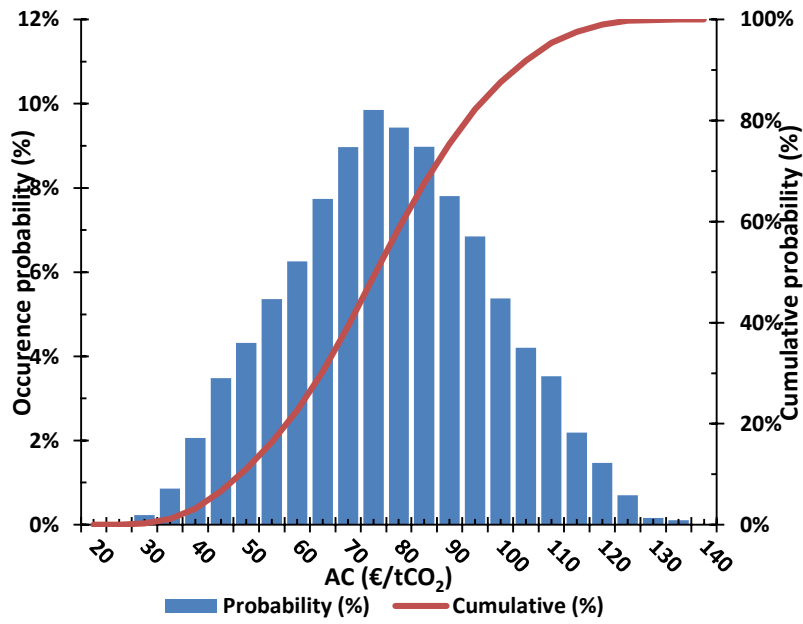


Figure 6-2 Occurrence and cumulative probability for a) levelized cost of electricity, b) cost of CO₂ avoided, c) cost of CO₂ captured of the biomass-fuelled oxy-fuel combustion power plant with the sCO₂ power cycle

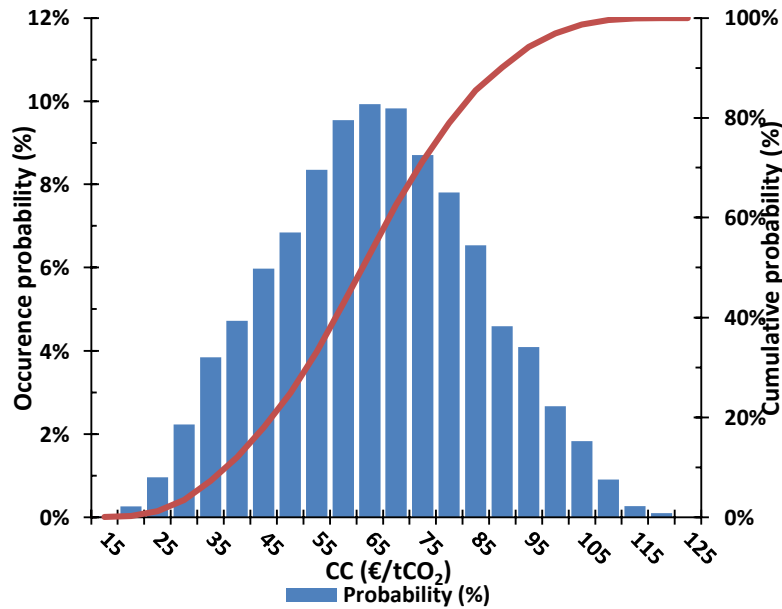
Figure 6-1, Figure 6-2 and Table 6-2 of the oxy-fuel combustion case reveal that LCOE, AC and CC of the coal-fuelled case are most likely in the ranges of 115-120 €/MWh (9.3%), 90-95 €/tCO₂ (6.9%), 55-60 €/tCO₂ (10.6%), respectively, and these of the biomass-fuelled case are most likely in these ranges of 100-120 €/MWh (9.4%), 20-30 €/tCO₂ (10.3%), 45-60 €/tCO₂ (9.6%), respectively. The mean value for LCOE, AC and CC of the coal-fuelled case are 111.7 €/MWh, 97.6 €/tCO₂, 52.8 €/tCO₂, respectively, and these of the biomass-fuelled case are 104.0 €/MWh, 29.2 €/tCO₂, 37.3 €/tCO₂, which means there is around 50% cumulative probability below each number. For the 5th and 95th percentiles, in the coal-fuelled case, LCOEs are estimated to be 73.7 €/MWh and 151.0 €/MWh, ACs are estimated to be 46.0 €/tCO₂ and 150.9 €/tCO₂, and CCs are estimated to be 19.7 €/tCO₂ and 86.9 €/tCO₂. Similarly, in the biomass-fuelled case, LCOE is estimated to be -56.6 €/MWh and 263.3 €/MWh, AC is estimated to be -43.7 €/tCO₂ and 101.5 €/tCO₂, and CC is estimated to be 80.2 €/tCO₂ and 153.7 €/tCO₂. In the kinds of literature, an oxy-coal combustion power plant with the sCO₂ power cycle

[216] achieved LCOE from 103.0 €/MWh to 116.6 €/MWh by changing CO₂ capture efficiency from 99% to 90%. In Table 6-1, the LCOE of 128.1 €/MWh and 133.7 €/MWh [217] were achieved in the oxy-coal combustion power plant with 593°C/24.1 MPa and 730°C/27.6 MPa. Comparing to an oxy-coal combustion power plant with a steam cycle [103], it achieved 117.5 €/MWh of LCOE. If considering the advantages at cost-saving of the sCO₂ power cycle, the LCOE of the oxy-coal combustion power plant with the sCO₂ power cycle is in the possible range of LCOE and going to be near the most probable range in Figure 6-1. From the above, it reveals interesting results that the most likely ranges and the mean value of LCOE, AC and CC in the coal-fuelled case are lower than those in the biomass-fuelled case. which by virtue to the carbon tax is considered. It proves, in the ETS, biomass-fuelled case has the advantage in terms of economy and decarbonisation in the market of solid-fuelled oxy-fuel power plants. Also, in terms of the results for the 5th and 95th percentiles, the biomass case demonstrates much lower LCOE in the 5th percentile by virtue to the higher carbon tax and much higher LCOE due to the higher fuel price. AC of the biomass case reaches the lowest value to -43.7 €/tCO₂ in the 5th percentile. CC of the biomass case has higher values than that of the coal case in terms of 5th and 95th percentiles. In addition, comparing Figure 6-1, Figure 6-2 and Table 6-2, the probability ranges of LCOE, AC and CC in the biomass case are wider due to the joint effect of the fuel price and carbon tax.



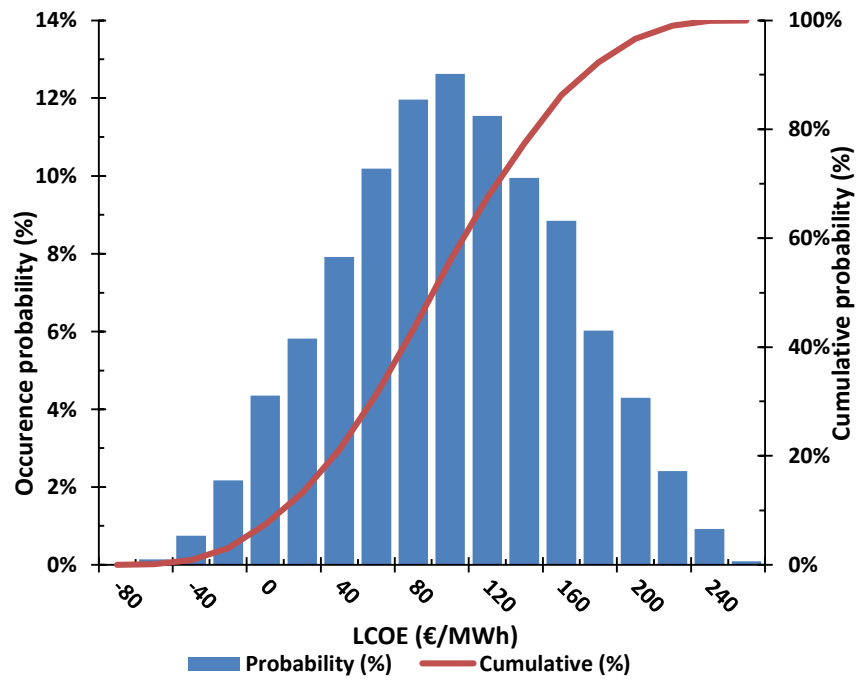


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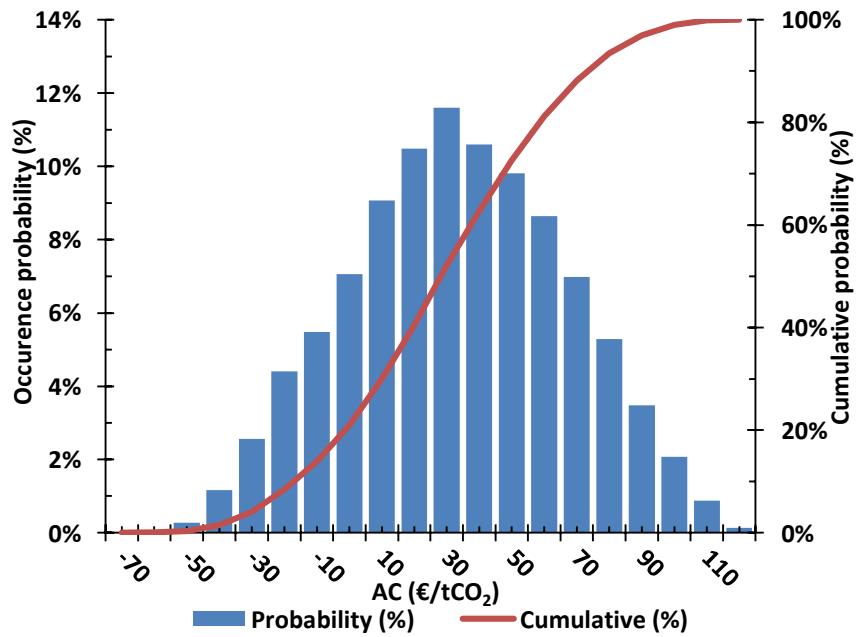


c)

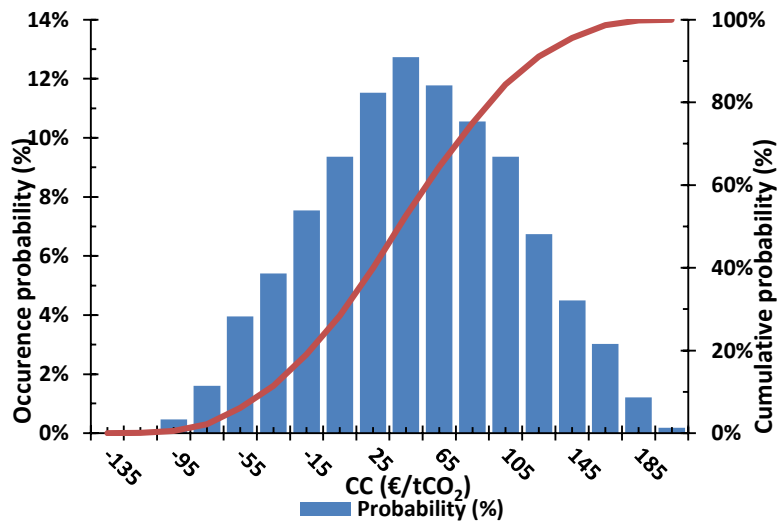
Figure 6-3 Occurrence and cumulative probability for a) levelised cost of electricity, b) cost of CO₂ avoided, c) cost of CO₂ captured of the coal-fuelled chemical looping combustion power plant with the sCO₂ power cycle



a)



b)



c)

Figure 6-4 Occurrence and cumulative probability for a) levelized cost of electricity, b) cost of CO₂ avoided, c) cost of CO₂ captured of the biomass-fuelled chemical looping combustion power plant with the sCO₂ power cycle

Figure 6-3, Figure 6-4 and Table 6-3 of the CLC case reveal that LCOE, AC and CC of the coal-fuelled case are most likely in the ranges of 95-100 €/MWh (12.2%), 75-80 €/tCO₂ (9.6%), 65-70 €/tCO₂ (9.8%), respectively, and that of the biomass-fuelled case are most likely in this ranges of 80-100 €/MWh (12.6%), 20-30 €/tCO₂ (11.4%), 25-45 €/tCO₂ (13.1%), respectively. The mean value of LCOE, AC and CC of the coal-fuelled case are 98.5 €/MWh, 75.9 €/tCO₂, 64.0 €/tCO₂, respectively, and these of the biomass-fuelled case are 90.8 €/MWh, 28.3 €/tCO₂, 41.1 €/tCO₂. From the above, comparing to the results of oxy-fuel combustion in the last paragraph, it can be concluded that the economic performance indicators of CLC cases present a sizable improvement apart from the CC increase essentially owing to the higher net electrical efficiency of CLC. Meanwhile, comparing with the oxy-fuel combustion cases, the statistical results of LCOE and AC regarding CLC have a relatively smaller standard deviation, which means the probability ranges of the LCOE and AC are smaller. In the coal-fuelled case, the figures for 5th and 95th percentiles of LCOE are estimated to be 67.9 €/MWh and

129.0 €/MWh, these of AC are estimated to be 36.4 €/tCO₂ and 115.4 €/tCO₂, and these of CC are estimated to be 26.0 €/tCO₂ and 101.9 €/tCO₂. Similarly, in biomass-fuelled cases, these of LCOE are predicted to be -30.2 €/MWh and 211.9 €/MWh, these of AC are estimated to be -38.8 €/tCO₂ and 95.4 €/tCO₂, and these of CC are estimated to be 79.7 €/tCO₂ and 161.8 €/tCO₂. Analysing the above results of LCOE, AC and CC, the CLC cases reached similar conclusions to oxy-fuel combustion cases in the last paragraph. Changing fuels and considering carbon taxes in the CLC have a significant impact on economic results. Also, in the literature in Table 6-1, LCOE of 121.1 €/MWh and 117.9 €/MWh is achieved and is in the range of the possible range of LCOE in Figure 6-3.

Table 6-1 Economic results with details of test cases [217]

Test Case	Technology	Plant size, MW	TIT, °C/MPa	LCOE, €/tCO ₂ (with CCS)	
				Steam cycle	sCO ₂ cycle
1	Oxy-coal	550	593/24.1	127.925	128.095
2	Oxy-coal		730/27.6	132.855	133.705
3	CLC		593/24.1	121.465	121.125
4	CLC		730/27.6	123.25	117.895
5	PC	90	593/24.1	107.1	136.425
6	PC		730/27.6	--	154.02

PC: pulverised coal power plant

It is interesting to note that cumulative probability curves of LCOE for considered cases were visually compared in Figure 6-5. In the coal-fuelled cases, it reveals that the CLC power plant with recompression sCO₂ power cycle has a higher cumulative probability at the same LCOE than the oxy-fuel combustion power plant with recuperated sCO₂ power cycle. However, in the biomass-fuelled cases, there is an intersection when LCOE is near 60 €/MWh. When the LCOE was below 60 €/MWh, the cumulative probability of LCOE for the oxy-fuel combustion power plant with recuperated sCO₂ power cycle is slightly higher than that of the CLC power plant with recompression sCO₂ power cycle.

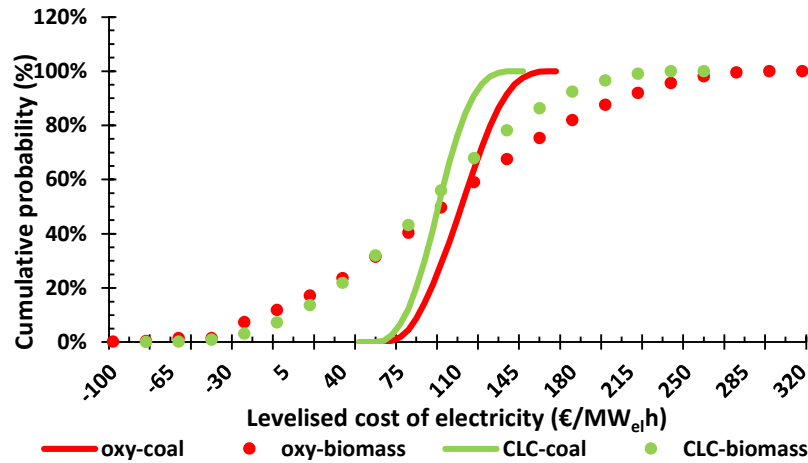


Figure 6-5 Cumulative probability of the two proposed systems fuelled by coal and biomass

Table 6-2 Key probability indicators for economic performance on coal- and biomass-fuelled oxy-combustion power plant with the recuperated sCO₂ power cycle

	Oxy-coal			Oxy-biomass		
	LCOE	AC	CC	LCOE	AC	CC
MIN	61.4	29.5	9.1	-96.3	-61.7	-109.2
MAX	166.0	171.2	99.9	305.6	120.7	184.7
MEAN	111.7	97.6	52.8	104.0	29.2	37.3
Std	19.6	26.6	17.1	80.0	36.3	58.5

Oxy-coal: oxy-fuel combustion with coal; Oxy-biomass: oxy-fuel combustion with biomass; MIN: minimum value; MAX: maximum value; MEAN: mean value; Std: standard deviation

Table 6-3 Key probability indicators for economic performance on coal- and biomass-fuelled chemical looping combustion power plant with the recompression sCO₂ power cycle

	CLC-coal			CLC-biomass		
	LCOE	AC	CC	LCOE	AC	CC
MIN	57.8	23.4	15.0	-81.0	-67.0	-127.4
MAX	143.0	133.5	120.8	250.1	116.7	204.6
MEAN	98.4	75.8	63.8	90.9	28.4	41.2
Std	15.3	19.7	19.0	60.6	33.6	60.4

CLC-coal: chemical looping combustion with coal; CLC-biomass: chemical looping combustion with biomass; MIN: minimum value; MAX: maximum value; MEAN: mean value; Std: standard deviation

7 General discussion

The main aim of this thesis was to explore opportunities for improving the techno-economic performance of the state-of-art oxy-fuel combustion power plant. Hence, two proposed systems were investigated with assessment methods including techno-economic assessment and probabilistic assessment. The first proposed system, the oxy-fuel combustion power plant with the recuperated sCO₂ power cycle, was assessed in Chapter 4. The second proposed system, the CLC power plant with recompression sCO₂ power cycle, was assessed in Chapter 5. The stochasticity of economic performance for the two projects was assessed in Chapter 6. It was found that employing a sCO₂ power cycle in an oxy-fuel combustion power plant can improve the techno-economic performance of oxy-fuel combustion and integrating the CLC and recompression sCO₂ power cycle achieved the best techno-economic performance in this thesis. From the series of assessments, it was confirmed that the sCO₂ power cycle and CLC are able to improve the techno-economic performance of the state-of-art oxy-fuel combustion power plants.

Table 7-1 presents a comparison of the techno-economic performance of the considered cases after unifying the key techno-economic assumptions [102], including the reference case of Case B12A [102], oxy-SC [103] oxy-sCO₂ and CLC-sCO₂. Case B12A is a state-of-art pulverized coal power plant without CO₂ capture and storage, and oxy-SC is a state-of-art oxy-fuel combustion power plant, which operates on similar temperature and pressure with the studied cases. With respect to Case B12A, oxy-SC shows a 10.2% net efficiency penalty and a 105% LCOE increase, which are the major challenges of oxy-combustion technology, and that is addressed in this work. Based on oxy-SC, oxy-sCO₂ fuelled by coal achieves 4.2% of further net efficiency penalty owing to the characteristic of the recuperated sCO₂ power cycle. It was found that its net power efficiency will surpass the oxy-SC by further increasing the TIT or the COP [218–220], such as

700°C and 300 bar. Its LCOE is mainly improved by virtue of the advantages of cost saving on the sCO₂ power cycle [218,219].

Table 7-1 Summary for the techno-economic performance of all studied cases

		Case B12A	oxy-SC	oxy-sCO ₂		CLC-sCO ₂ (Mn-ore)	
		Coal	Coal	Coal	Biomass	Coal	Biomass
Fuel		Coal	Coal	Coal	Biomass	Coal	Biomass
Temperature	°C	593	599	593	593	593	593
Pressure	MPa	24.1	24.1	24.1	24.1	24.1	24.1
Chemical energy input	MJ/s	1613.9	1830.1	1633.0	1633.0	1566.0	1566.0
Gross power output	MW _{el}	685	783.5	615.9	587.0	649.4	599.4
Auxiliary consumption	MW _{el}	35	233.3	193.3	200.6	93.6	102.2
Net power output	MW _{el}	650	550.2	422.6	386.4	555.8	497.2
Net efficiency (HHV)	%	40.3%	30.1%	25.9%	23.7%	35.5%	31.7%
Specific capital cost	€/kW _{el}	1868.1 ^b	3207.6 ^a	2650.9 ^c	2810.2 ^c	2790.7 ^c	3084.4 ^c
Capital COE	€/MW _{elh}	25.2	65.5	41.3	43.8	46.6	51.5
Fixed O&M COE	€/MW _{elh}	8.5	14.1	3.6	3.8	11.1	11.0
Variable O&M COE w TS&M	€/MW _{elh}	6.9	8.2	18.3	20.9	13.9	16.3
Fuel COE	€/MW _{elh}	16.8	29.7	34.5	68.5	25.2	51.0
LCOE	€/MW _{elh}	57.3 ^b	117.5 ^a	97.7 ^c	136.8 ^c	96.8 ^c	129.8 ^c

SC: steam Rankine cycle

a: base year of 2011

b: base year of 2018

c: base year of 2019

Regarding CLC-sCO₂ fuelled by coal, compared with the oxy-SC case, its net efficiency and LCOE achieve 5.4% and 20.7 €/MW_{elh} improvements, mainly by virtue of the lower energy penalty of the CLC power plant versus ASU in the oxy-fuel combustion power plant [221] and higher cycle efficiency of the recompression sCO₂ power cycle versus the recuperated sCO₂ power cycle which are comparable to previous results [218,220,222,223]. It should be noticed that there is an optimal value of COP (20-24 MPa) on the plant net efficiency in TIT ranging from 550 to 800°C [223]. The unit cost of the OCs largely affects the overall economic performance. Regarding the biomass-fuelled cases of the two-generation oxy-fuel combustion power plants, each case has a carbon tax to determine the type of fuel, which provides a reference for countries. As in the literature [70,224], the feasibility of technology implementation is able to be determined by the carbon price.

Moreover, in the stochastic economic assessment, the CLC power plant with the recompression sCO₂ power cycle shows better performance. In common with the literature [225], in coal-fuelled cases, selection of the OC and complexity of the dual bed operation are regarded as the potential issues influencing LCOE. In

biomass-fuelled cases, the carbon tax is an additional consideration for the LCOE. It is worth noting that, in this thesis, it is challenging to compare the technologies at a high technical readiness level with those at a lower technical readiness level. From the uncertainty range of the economic indicators, it is noncommittal to choose and exploit a satisfactory technology, but further research and development are reasonable and necessary to reduce the overall error bounds.

In the initial feasibility study phase of the project, simulation is a good tool to estimate the overall cost, such as Aspen Plus utilized in this thesis. In reality, the continuous maturity of technology means that the scale of the project is constantly expanding (from pilot scale to demonstration scale, until commercial scale). According to the theory of economies of scale, the unit production cost of a project (such as LCOE) decreases as the scale increases.

In summary, to achieve the described aim of the thesis, technologies of CLC and sCO₂ power cycle, as the alternative ways for the state-of-art oxy-fuel combustion power plant, achieve outstanding techno-economic performance through identification, quantification and analysis. Meanwhile, the biomass-fuelled cases prove that they are able to surpass the coal-fuelled cases under the ETS with excellent decarbonization ability.

8 General conclusion

8.1 Oxy-fuel combustion with the recuperated supercritical CO₂ power cycle

This study presented an investigation of the techno-economic performance of the novel oxy-combustion coal- and biomass-fired power plant with the recuperated sCO₂ cycle and compared this with the reference oxy-combustion CFPP with the conventional steam cycle. This study has proved that:

- Replacement of the conventional steam cycle (SC-coal case) with the sCO₂ cycle (sCO₂-coal case) resulted in a reduction in the net efficiency by 0.4%_{HHV} and an increase in the LCOE by 0.6 €/MW_{el}h;
- Replacement of the fuel from coal (sCO₂-coal case) to wood pellets (sCO₂-WP case) resulted in a further reduction in the net efficiency of 2.3%_{HHV} and an increase in the LCOE of 31.2 €/MW_{el}h but enabled achieving a substantial negative CO₂ emission of 1255.2 gCO₂/kW_{el}h.
- Compared to the reference case, the sCO₂-WP case had better environmental performance, but had poorer thermodynamic and economic performance under the initial conditions with no carbon tax considered. Therefore, the deployment of BECCS relies on the development of an efficient CO₂ market that will support carbon-negative technologies.
- Potential improvements in the techno-economic performance are achievable by increasing the TIT and COP. Both parameters were positively correlated with the net efficiency and negatively correlated with the LCOE and AC. Moreover, fuel prices and discount rates were shown to be positively correlated with LCOE and AC.
- The sCO₂-coal case will result in a 2%_{HHV} points higher net efficiency and 4.1 €/MW_{el}h lower LCOE than that for the reference SC-coal case. As this study utilized a simple recuperated sCO₂ cycle, such outcome confirms that implementation of advanced power cycles can substantially reduce the economic and efficiency penalties associated with CCS; and

- The inherent ability of the sCO₂-WP case to operate with negative CO₂ emissions was shown to offset the increased efficiency penalty when the carbon tax was above 23.18 €/tCO₂. Above this value of the carbon tax, the sCO₂-WP case became more economically feasible than the sCO₂-coal case. Moreover, if the carbon tax was above 95.70 €/tCO₂, the revenue associated with offsetting the CO₂ emissions resulted in an LCOE below zero.

8.2 Chemical looping combustion with recompression supercritical CO₂ power cycle

This study presented an investigation of the techno-economic performance of the novel CLC coal- and biomass-fired power plant with the recompression sCO₂ cycle and compared this with the reference chemical looping combustion CFPP with the conventional steam cycle. This study has proved that:

- The natural OC based CLC power plant achieved better techno-economic performance than the manufactured OC based, such as the Manganese ore as the OC performed best in this work;
- Natural OC materials, such as limestone and ilmenite, result in the energy penalty owing to their endothermic decomposition reaction in the fuel reactor.
- Replacement of the conventional steam cycle with the sCO₂ cycle and additional flue gas recycle to the fuel reactor resulted in an increase in the net efficiency by 0.43%_{HHV} and a reduction in the LCOE by 1.4 €/MW_{elh}. Further consideration of the Manganese ore as the OC achieved an increase in the net efficiency by 0.42%_{HHV} and a reduction in the LCOE by 12.4 €/MW_{elh}.
- Further replacement of the fuel from coal to wood pellets in the Mn₃O₄ case resulted in a further reduction in the net efficiency of 3.32%_{HHV} and an increase in the LCOE of 20.6 €/MW_{elh}, but achieved a substantial negative CO₂ emission of 1066 gCO₂/kW_{elh}.
- Compared to the reference case, the biomass-fuelled cases had better environmental performance, but had poorer thermodynamic and economic performance under the initial conditions without carbon tax considered.

Therefore, the deployment of BECCS relies on the development of an efficient CO₂ market, such as the EU ETS, that will support the biomass-fuelled CLC power plant with a sCO₂ power cycle.

- Under the same TIT, the highest net electricity efficiency is when the COP around 200 bars, which results in the lower LCOE and AC. Moreover, discount rate, CO₂ TS&M unit cost, OC prices and fuel prices were shown to be positively correlated with techno-economic performance, apart from the CO₂ TS&M unit cost did not impact the AC. And the capacity factor negatively impacts the techno-economic performance.
- The techno-economic performance deteriorated on the increase in the OC attrition rate.
- The relatively small net efficiency penalty resulting from the char conversion efficiency did not hold back the decrease of AC and CC by virtue to the inherent decarbonisation ability of the CLC power plant.
- The inherent negative emission ability of the biomass-fuelled case using manganese ore was shown to offset the increased efficiency penalty when the carbon tax was above 30.1 €/tCO₂. Above this value, the biomass-fuelled case became more economically feasible than the coal-fuelled. If the carbon tax was above 121.8 €/tCO₂, the revenue associated with decarbonisation resulted in an LCOE below zero. Moreover, when the carbon tax is over 16.6 €/tCO₂ the revenue of decarbonisation leads to a cheaper CC than the coal-fuelled case.

8.3 Probability economic assessment

This study presented an investigation of the economic probability of the two proposed power plants. One is the oxy-fuel combustion power plant with a recuperated sCO₂ power cycle. Another one is the CLC power plant with recompression sCO₂ cycle). This study has proved that:

- The statistical distribution for LCOE, AC and CC is close to the normal distribution.

- The most likely ranges and the mean value of LCOE, AC and CC in the coal-fuelled case are lower than those in the biomass-fuelled case, which by virtue to the carbon tax is considered. It demonstrates, in the ETS, biomass-fuelled case has the advantage in terms of economy and decarbonisation in the market of solid-fuelled oxy-fuel power plants.
- Regarding the results of the 5th and 95th percentiles, the biomass case demonstrates much lower LCOE in the 5th percentile by virtue to the higher carbon tax and much higher LCOE due to the higher fuel price. AC of the biomass case reaches the lowest value in the 5th percentile. CC of the biomass case has higher values than that of the coal case in terms of 5th and 95th percentiles.
- The probability ranges of LCOE, AC and CC in the biomass case are wider due to the joint effect of the fuel price and carbon tax.
- In terms of the cumulative probability of LCOE, the coal-fuelled CLC has better performance than the oxy-coal combustion case. After changing the fuel to biomass, the CLC has a higher cumulative probability than the oxy-fuel combustion when the LCOE is over around 60 €/MWh.

8.4 Future work

The sCO₂ power cycle is considered for improving the first and second generation oxy-fuel combustion in this thesis. However, its TRL is still lower due to the technical uncertainty of the key facilities, which is necessary to be studied in future work. For example, the sCO₂ turbomachinery has various knowledge and technology gaps, and the design of the cooler must consider the CO₂ near the critical region. In addition, consideration for the material of sCO₂ system facilities is required due to the higher temperature and pressure, large variation of temperature and pressure, and corrosion of CO₂. The control system for the sCO₂ power system has not been extensively and deeply studied. In terms of the CLC, significant progress is still needed in the mechanism of redox reactions, catalyst design and optimization, reactor and process design and operation, and process

amplification of the existing CLC system. Many studies investigated different CLC processes, such as chemical looping solid oxide fuel cell and chemical looping air separation [226]. The sCO₂ power cycle, too, has been explored by plenty of experiments and research. In addition, from the thermodynamic point of view, CLC and the sCO₂ power cycle are more advantageous than the first generation oxy-fuel combustion technology and conventional steam Rankine cycle. Further reducing their cost will highlight their overall advantage. By referring to the theory of economies of scale, upgrading the level (capacity) of the power plant can further reduce the overall cost. Therefore, the investigation and research of demonstration power plants are conducive to clarify the techno-economic performance for various scales and provide a theoretical basis for the future commercial and utility-scale.

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Appendix A Aspen Plus™ Physical Property Databanks and block categories

Table A-1 Aspen Plus™ Physical Property Databanks [227]

Databank Name	Password	Type	Maximum Parameter	Maximum Components	Maximum Pairs	Description
PURE11	PURE11	PP1	100	2500	—	Main pure component databank
ASPENPCD	ASPENPCD	PP1	40	1000	—	Aspen Physical Property System pure component databank
SOLIDS	SOLIDS	PP1	40	4000	—	Solid component databank
AQUEOUS	AQUEOUS	PP1	40	4000	—	Aqueous component databank
BINARY	BINARY	PP2	20	100	3000	Binary databank
COMBUST	COMBUST	PP1	40	4000	—	Combustion databank
ETHYLENE	ETHYLENE	PP2	20	100	3000	Ethylene databank
INORGANIC	INORGANIC	PP1	25	2500	—	Inorganic component databank
PURE856	PURE856	PP1	100	2500	—	Pure component databank from release 8.5 – 6
PURE93	PURE93	PP1	100	2500	—	Pure component databank from release 9.3
PURE10	PURE10	PP1	100	2500	—	Pure component databank from version 10.2
AQU92	AQU92	PP1	40	4000		AQUEOUS databank for Release 9.2

Table A-2 Aspen Plus™ block categories [228]

Type	Model	Description
Mixers/Splitters	Mixer	Stream mixer
	FSplit	Stream splitter
	SSplit	Substream splitter
Separators	Flash2	Two-outlet flash
	Flash3	Three-outlet flash
	Decanter	Liquid-liquid decanter
	Sep	Multi outlet component separator
	Sep2	Two-outlet component separator
Heat Exchangers	Heater	Heater/cooler
	HeatX	Two-stream heat exchanger
	MheatX	Multistream heat exchanger
	HxFlux	Interface to BJAC Shell & Tube heat exchanger program
	Hetran	Interface to BJAC air cooled heat exchanger program
	Aerotran	Interface to the Aspen Aerotran program
Columns	HTRIIST	Interface to the IST program
	DSTWU	Shortcut distillation design
	Distl	Shortcut distillation rating
	RadFrac	Rigorous distillation
	Extract	Rigorous liquid-liquid extractor
	MultiFrac	Rigorous distillation for complex columns
	SCFrac	Shortcut distillation for petroleum
	PetroFrac	Rigorous distillation for petroleum
	RateFrac	Rate-based distillation
	BatchFrac	Rigorous batch distillation
Reactors	RStoic	Stoichiometric reactor
	RYield	Yield reactor
	REquil	Equilibrium reactor
	RGibbs	Equilibrium reactor
	RCSTR	Continuous-stirred tank reactor
	RPlug	Plug flow reactor

Pressure Changers	RBatch	Batch reactor
	Pump	Pump/hydraulic turbine
	Compr	Compressor/turbine
	MCompr	Multistage compressor/turbine
	Pipeline	Multi segment pipeline pressure drop
	Pipe	Single segment pipeline pressure drop
	Valve	Rigorous valve pressure drop
Manipulators	Mult	Stream multiplier
	Dupl	Stream duplicator
	ClChng	Stream class changer
	Selector	Stream selector
Solids	Crystallizer	Mixed suspension mixed product removal crystallizer
	Crusher	Solids crusher
	Screen	Solids separator
	FabFI	Fabric filter
	Cyclone	Cyclone separator
	VScrub	Venturi scrubber
	ESP	Electrostatic precipitator
	HyCyc	Hydrocyclone
	CFuge	Centrifuge filter
	Filter	Rotary vacuum filter
	Swash	Single-stage solids washer
	CCD	Counter-current decanter
User models	User, User2	User-supplied Fortran unit operation models
	Excel Spreadsheets	Excel spreadsheets interfaced through User2
	ACM flowsheets	Flowsheet exported from ACM or AD
	CAPE-OPEN unit operation	COM unit operations developed on VB or C++
	Hierarchy	Hierarchical structure

Appendix B Data from thermodynamic assessment required by economic assessment in the case of oxy-fuel combustion with sCO₂ power cycle and chemical looping combustion with sCO₂ power cycle

Table B-1 Data from thermodynamic assessment required by economic assessment in the case of oxy-fuel combustion with sCO₂ power cycle

Component	Parameter	Unit
	Gross electric capacity	MW _{el}
	Auxiliary power consumption	MW _{el}
	Net power output	MW _{el}
	Net efficiency	%
	Enhancer Gas Recycle mass flow rate	kg/s
	OC circulation flow rate (ideal)	kg/s
	OC circulation flow rate (real)	kg/s
	OC makeup flow rate	kg/s
	Ash out	kg/s
	Air in	kg/s
	Total induced draft fan passed gas	kg/s
	CO ₂ mass flow rate in CO ₂ product	kg/s
	CO ₂ emission rate	kg/s
	CO ₂ in air	kg/s
	N ₂ in CO ₂ product	kg/s
	SO ₂ in CO ₂ product	kg/s
Main compressor and/or recompressor	Power consumption	MW _{el}
Condenser	Duty	MW _{el}
Forced Draft Fans	Power consumption	MW _{el}
Induced Draft Fans	Power consumption	MW _{el}
Enhancer Gas Recycle Compressors	Power consumption	MW _{el}
CO ₂ Compressor	Power consumption	MW _{el}
Main compressor	CO ₂ flow rate	kg/s
	Outlet pressure	bar
Recompressor	CO ₂ flow rate	kg/s
	Outlet pressure	bar
Expander	CO ₂ flow rate	kg/s
	Inlet temperature	°C
	Inlet pressure	bar
Generator	Power	kW
HTR	Highest pressure	bar
	Load capacity	kW
	LMTD	°C

LTR	Load capacity	kW
	LMTD	°C
Preheater	Highest pressure	bar
	Load capacity	kW
	LMTD	°C
FR HX	Highest pressure	bar
	Load capacity	kW
	Hot inlet temperature	°C
	Cool inlet temperature	°C
	Cool outlet temperature	°C
AR HX	Highest pressure	bar
	Load capacity	kW
	Hot inlet temperature	°C
Cooler	Highest pressure	bar
	Load capacity	kW
	Hot inlet temperature	°C
	Hot outlet temperature	°C

LMTD: Logarithmic mean temperature difference

Table B-2 Data from thermodynamic assessment required by economic assessment in the case of chemical looping combustion with sCO₂ power cycle

Component	Parameter	Unit
	Gross electric capacity	MW _{el}
	Net efficiency	%
Turbine	Inlet temperature	°C
Compressor	Outlet pressure	bar
Forced draft fans	Power consumption	MW _{el}
Induced draft fans	Power consumption	MW _{el}
CPU	Power consumption	MW _{el}
Compressor	Flow rate	kg/s
Expander	Flow rate	kg/s
	Highest temperature	k
	Highest pressure	bar
Generator	Power	kW
Recuperator	Highest pressure	bar
	Load capacity	W
	Hot inlet temperature	K
	Hot outlet temperature	K
	Cool inlet temperature	K
	Cool outlet temperature	K
Primary heater	Highest pressure	bar
	Load capacity	W
	Hot inlet temperature	K
	Hot outlet temperature	K
	Cool inlet temperature	K
	Cool outlet temperature	K
Economiser	Highest pressure	bar
	Load capacity	W
	Hot inlet temperature	K
	Hot outlet temperature	K
	Cool inlet temperature	K
	Cool outlet temperature	K
Cooler	Load capacity	W
	Hot inlet temperature	K

Appendix C Summary of the techno-economic performance of chemical looping combustion power plants

Table C-1 Summary of the technical performance of chemical looping combustion power plants

		Fe ₂ O ₃			NiO		CaSO ₄	Mn ₃ O ₄		Fe ₂ TiO ₅	
Parameter	Unit	Reference	Coal	WP	Coal	WP	Coal	Coal	WP	Coal	WP
System input data											
Power cycle operation condition						593°C & 240 bar					
Fuel chemical energy input	MW _{th}	1566.0	1566.0	1566.0	1566.0	1566.0	1566.0	1566.0	1566.0	1566.0	1566.0
Fuel higher heating value	kJ/kg	27135.0	27135.0	16804.9	27135.0	16804.9	27135.0	27135.0	16804.9	27135.0	16804.9
Fuel feed rate	kg/s	57.7	57.7	93.2	57.7	93.2	57.7	57.7	93.2	57.7	93.2
Fixed char in fuel	kg/s	36.8	36.8	41.4	36.8	41.4	36.8	36.8	41.4	36.8	41.4
Air flow rate	kg/s	587.8	587.0	587.0	587.0	587.0	587.0	587.0	587.0	587.0	587.0
Enhancer Gas Recycle flow rate	kg/s	-	153.2	153.2	153.2	153.2	153.2	153.2	153.2	153.2	153.2
OC circulation flow rate	kg/s	11840.6	11840.6	11891.0	1636.5	1636.7	767.9	2442.4	2442.4	3999.9	4000.0
OC makeup flow rate	kg/s	1.3	1.3	1.3	0.4	0.4	4.5	2.4	2.4	16.0	16.0
Auxiliary electricity requirement of CLC-sCO₂ power plant											
Coal handling and conveying	MW _{el}	0.5	0.5	0.8	0.5	0.7	0.4	0.4	0.7	0.4	0.7
Pulverisers	MW _{el}	3.1	3.1	5.0	3.1	5.0	3.1	3.1	5.0	3.1	5.0
Coal injection system	MW _{el}	1.0	1.0	1.6	1.0	1.6	1.0	1.0	1.6	1.0	1.7
Sorbent handling & reagent preparation	MW _{el}	0.8	0.8	1.2	0.8	1.2	0.8	0.8	1.2	0.8	1.2
Carrier particle handling	MW _{el}	0.8	1.8	1.8	0.5	0.5	1.0	0.6	0.6	3.8	3.8
Ash handling	MW _{el}	0.7	0.7	0.1	0.7	0.1	0.7	0.7	0.1	0.7	0.1
Forced draft fans	MW _{el}	6.5	11.7	11.7	12.2	11.7	11.7	12.2	11.7	12.2	11.7
Induced draft fans	MW _{el}	3.9	8.1	9.1	8.4	9.1	8.2	8.4	9.1	8.4	9.1
SCR	MW _{el}	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
Baghouse	MW _{el}	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Wet FGD	MW _{el}	6.4	2.1	2.1	2.2	2.1	2.1	2.2	2.1	2.2	2.1
Enhancer gas recycle compressors	MW _{el}	-	7.2	7.2	7.2	7.2	7.3	7.2	7.2	7.2	7.3
HCl scrubber pump	MW _{el}	0.04	0.04	-	0.04	-	0.04	0.04	-	0.04	-
CO ₂ compressor	MW _{el}	55.9	48.0	53.0	46.6	51.4	49.2	48.5	53.5	48.0	53.0
Miscellaneous balance of plant	MW _{el}	2.0	2.0	3.2	2.0	3.2	2.0	2.0	3.2	2.0	3.2
Steam turbine auxiliaries	MW _{el}	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4	0.4

Parameter	Unit	Fe ₂ O ₃			NiO		CaSO ₄	Mn ₃ O ₄		Fe ₂ TiO ₅	
		Reference	Coal	WP	Coal	WP	Coal	Coal	WP	Coal	WP
Condensate pumps	MW _{el}	0.9	-	-	-	-	-	-	-	-	-
Circulating water pumps	MW _{el}	5.0	-	-	-	-	-	-	-	-	-
Cooling tower fans	MW _{el}	2.6	4.1	3.8	4.1	3.8	4.0	4.1	3.8	4.1	3.7
Transformer losses	MW _{el}	2.2	2.0	1.9	2.0	1.9	2.0	2.0	1.9	2.0	1.9
Total	MW_{el}	92.7	93.6	103.0	91.8	100.1	94.0	93.6	102.2	96.3	104.9
System technical performance indicators											
Main compressor and recompressor	MW _{el}	-	436.8	403.1	434.5	400.4	425.8	436.0	402.4	432.1	398.5
Gross electricity production	MW _{el, gross}	641.8	649.4	599.3	647.2	596.4	634.2	649.4	599.4	643.6	593.5
Net electricity production	MW _{el, net}	549.1	555.8	496.3	555.4	496.3	540.2	555.8	497.2	547.2	488.6
Gross electric efficiency	% _{HHV}	40.98%	41.47%	38.27%	41.33%	38.08%	40.50%	41.47%	38.28%	41.10%	37.90%
Net electric efficiency	% _{HHV}	35.07%	35.49%	31.69%	35.47%	31.69%	34.50%	35.49%	31.75%	34.95%	31.20%
Net energy penalty	% _{HHV}	0	-0.43%	3.37%	-0.40%	3.37%	0.57%	-0.42%	3.32%	0.12%	3.87%
	MW _{el, net}	0	-6.66	52.80	-6.34	52.83	8.93	-6.65	51.92	1.86	60.55
Specific fuel chemical energy consumption	kW _{th} /kW _{el, net}	2.9	2.8	3.2	2.8	3.2	2.9	2.8	3.1	2.9	3.2
Specific fuel consumption	g _{fuel} /kW _{el, net}	378.4	373.8	675.9	374.0	676.0	384.6	373.8	674.7	379.6	686.6
CO ₂ product purity	wt%	99.22%	99.29%	99.83%	99.26%	99.69%	99.32%	99.30%	99.83%	99.29%	99.82%
Annual CO ₂ captured rate	Mton/yr	3.46	3.47	3.91	3.33	3.75	3.42	3.50	3.95	3.47	3.90
Annual CO ₂ emission rate	Mton/yr	0.13	0.16	-3.9	0.28	-3.7	0.16	0.12	-3.9	0.16	-3.9
CO ₂ emission per fuel energy input	kgCO ₂ /GJ	3.6	3.79	-93.04	6.56	-89.25	3.79	2.94	-94.01	3.80	-92.96
specific CO ₂ emission	gCO ₂ /kW _{el, net}	36.9	38.4	-1,057	66.6	-1,014	39.6	29.9	-1,066	39.2	-1,073
specific CO ₂ captured	gCO ₂ /kW _{el, net}	848.9	838.1	1056.8	804.5	1013.9	850.7	846.9	1065.9	850.5	1072.7
CO ₂ capture efficiency	wt%	95.80%	95.61%	95.66%	91.70%	91.76%	94.33%	96.59%	96.65%	95.51%	95.57%

Table C-2 Summary of the economic performance of chemical looping combustion power plants

Parameter	Unit	Fe ₂ O ₃			NiO		CaSO ₄	Mn ₃ O ₄		Fe ₂ TiO ₅	
		Reference	Coal	WP	Coal	WP	Coal	Coal	WP	Coal	WP
Power cycle operation condition											
System economic performance											
593°C & 240 bar											
TCR in reference cases	M€ (2011)	1456.3	-	-	-	-	-	-	-	-	-
1). steam turbine	M€	143.0	-	-	-	-	-	-	-	-	-
2). Feedwater & Misc BOP Systems	M€	88.6	-	-	-	-	-	-	-	-	-
3). HRSG	M€	59.7	-	-	-	-	-	-	-	-	-
TCR w/o 1), 2), 3)	M€ (2011)	1165.0	-	-	-	-	-	-	-	-	-
	M€ (2019)	1208.3	-	-	-	-	-	-	-	-	-
sCO ₂ power cycle											
Bare Erected Cost	M€	-	137.1	126.9	136.2	125.9	133.7	136.6	126.5	135.4	125.3
Compressors	M€	-	1.6	1.5	1.6	1.5	1.5	1.6	1.5	1.6	1.5
Expander	M€	-	89.1	82.3	88.7	81.7	87.2	89.0	82.1	88.2	81.3
Heat exchangers	M€	-	10.9	10.3	10.6	10.0	10.4	10.6	10.1	10.6	10.0
Electric generators	M€	-	28.9	26.8	28.8	26.7	28.3	28.9	26.8	28.7	26.6
Piping	M€	-	6.5	6.0	6.5	6.0	6.4	6.5	6.0	6.4	6.0
Eng'g CM H.O. & Fees	M€	-	12.0	11.1	12.0	11.1	11.7	12.0	11.1	11.9	11.0
Process Cont.	M€	-	20.6	19.0	20.4	18.9	20.1	20.5	19.0	20.3	18.8
Project Cont.	M€	-	25.5	23.6	25.3	23.4	24.8	25.4	23.5	25.1	23.3
Owner's cost	M€	-	41.0	37.9	40.7	37.6	40.0	40.8	37.8	40.5	37.5
sCO ₂ power cycle in TRC	M€	-	236.1	218.6	234.6	216.8	230.4	235.3	217.9	233.3	215.8
	€/kW _{el, net}	-	424.8	440.4	422.3	437.0	426.4	423.5	438.3	426.3	441.8
TRC	M€	1456.3	1444.4	1426.9	1442.9	1425.2	1546.0	1551.0	1533.5	1548.9	1531.5
Specific capital cost	€/kW _{el, net}	2,647.1	2,599.0	2,875.0	2,597.8	2,871.7	2,862.0	2,790.7	3,084.4	2,830.4	3,134.7
Annual FOM	M€/year	42.9	42.5	42.0	42.4	41.9	45.4	45.6	45.1	45.5	45.0
Annual VOM	M€/year	119.4	119.4	122.1	125.2	127.8	54.4	57.6	60.4	93.7	96.4
Annual fuel cost	M€/year	104.1	104.1	188.9	104.1	188.9	104.1	104.1	188.9	104.1	188.9
Capital COE	€/MW _{elh}	44.2	43.4	48.0	43.4	47.9	47.8	46.6	51.5	47.3	52.3
Fixed O&M COE	€/MW _{elh}	10.5	10.4	10.3	10.4	10.2	11.1	11.1	11.0	11.1	11.0
Variable O&M COE	€/MW _{elh}	29.2	28.9	33.1	30.3	34.6	13.5	13.9	16.3	23.0	26.5
Carbon tax COE	€/MW _{elh}	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
CO ₂ TS&M COE	€/MW _{elh}	5.9	5.9	7.4	5.6	7.1	6.0	5.9	7.5	6.0	7.5
OC COE	€/MW _{elh}	17.0	16.8	18.9	18.7	20.9	1.0	1.6	1.8	10.5	11.8

Parameter	Unit	Fe ₂ O ₃			NiO		CaSO ₄	Mn ₃ O ₄		Fe ₂ TiO ₅	
		Reference	Coal	WP	Coal	WP	Coal	Coal	WP	Coal	WP
Fuel COE	€/MW _{el} h	25.4	25.2	51.1	25.2	51.1	25.9	25.2	51.0	25.5	51.9
LCOE w/o TS&M (2019)	€/MW _{el} h	103.3	101.9	135.0	103.5	136.8	92.3	90.9	122.4	96.4	130.0
LCOE w TS&M (2019)	€/MW _{el} h	109.2	107.8	142.4	109.2	143.9	98.3	96.8	129.8	106.9	141.8
Reduction of LCOE	%	-	1.3	-30.4	0.1	-31.7	10.0	11.4	-18.9	2.1	-29.8
AC	€/tCO ₂	90.9	89.2	55.3	94.5	57.4	76.8	74.0	48.3	88.1	54.4
CC	€/tCO ₂	74.9	74.2	90.2	79.3	95.8	61.8	60.4	77.6	66.6	84.1

