



Decarbonising bioenergy through biomass utilisation in chemical looping combustion and gasification: a review

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Abstract

Biomass valorisation for bioenergy is crucial for establishing a sustainable low-carbon circular bioeconomy and addressing societal and environmental challenges. As global demand for renewable energy grows, effective waste management using biomass becomes increasingly vital. However, successful implementation requires addressing specific local, scientific, and conceptual challenges, including technological intricacies, resource availability, scalability, and environmental impacts. Advancements in chemical looping gasification and chemical looping combustion are reviewed here as essential components of bioenergy with carbon capture and storage technologies. Chemical looping technologies have substantially contributed to decarbonising various industries, supported by precise trends. The review highlights the potential of chemical looping combustion and gasification in biomass utilisation for generating bioenergy and mitigating carbon emissions. Moreover, the advantages of chemical looping processes enhance their feasibility and applicability across different contexts including (1) significant increase in biomass conversion efficiency through chemical looping gasification, leading to substantial reductions in greenhouse gas emissions, (2) progress in chemical looping combustion, enhancing carbon capture and storage capabilities and contributing to a closed carbon cycle, (3) detailed insights into integrating chemical looping technologies into industrial sectors.

Keywords Chemical looping combustion · Advanced combustion · Chemical looping gasification · Biomass · Biochar · Biomass valorisation

Abbreviations

ASR	Automotive shredder residue
BA	Bottom ash
BECCS	Bioenergy with carbon capture and storage
CLC	Chemical looping combustion
CLG	Chemical looping gasification
CLOU	Chemical looping oxygen uncoupling,
IGCC	Integrated gasification combined cycle
kWth	Kilowatt thermal
LD	Linz–Donawitz process

MWth	Megawatt thermal
OC	Oxygen carriers or metal oxides
OcBR	Oxygen carrier-to-biomass ratio
OBR	Oxygen-to-biomass ratio
SBR	Steam-to-biomass ratio
TGA	Thermogravimetric analyser

Chemical formulas

Al ₂ O ₃	Aluminium oxide
CaO	Calcium oxide
CH ₄	Methane
CO	Carbon monoxide
CO ₂	Carbon dioxide
Co ₃ O ₄	Cobalt(II, III) oxide
CoO	Cobalt(II) oxide
Co	Cobalt
CuO	Copper(II) oxide
Cu	Copper
Fe	Iron
Fe ₂ O ₃	Iron(III) oxide
H ₂	Hydrogen
H ₂ O	Steam or water

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K_2CO_3	Potassium carbonate
KCl	Potassium chloride
KOH	Potassium hydroxide
Li_2CO_3	Lithium carbonate
$MgAl_2O_4$	Magnesium aluminate
MgO	Magnesium oxide
Mn_2O_3	Manganese(III) oxide
Mn_3O_4	Manganese(II,III) oxide
MnO	Manganese(II) oxide
M_xO_y	Oxidised metal oxide
M_xO_{y-1}	Reduced metal oxide
Na_2CO_3	Sodium carbonate
NaCl	Sodium chloride
NaOH	Sodium Hydroxide
NiO	Nickel(II) oxide
N_2	Nitrogen
O_2	Oxygen
SiO_2	Silicon dioxide
ZrO_2	Zirconium dioxide

Greek letters

ΔG	Gibbs free energy
Q_{heat}	Heat produced via metal oxide oxidation

Notations

C_n	The number of carbon atoms in the fuel
h	Hour
H_p	The number of hydrogen atoms in the fuel
Me	Represent metal atom in metal oxide
Me_x	The number of metal atoms in metal oxide
O_m	The number of oxygen atoms in the fuel
O_y	The number of oxygen atoms in metal oxide

Introduction

Biomass is one of the major most valuable renewable energy sources, and it is important to the global carbon cycle (Shrivastava et al. 2021; Güleç et al. 2022d). Biomass can be used as a clean, environmentally friendly, and inexpensive precursor for energy and green chemicals production (Kumar et al. 2018; Kostas et al. 2019). Biomass is a carbon neutral source of fuel that is abundantly available and has the potential to complement petroleum-based fuels existing for heat and electricity generation (Andersen et al. 2021). As such, biomass conversion and utilisation processes could play a major role in achieving the new European Union net zero energy targets. In 2019, more than half of the European Union's renewable energy is produced from biomass based resources. It is expected that biomass contribution towards renewable energy production would continue to increase each year (Andersen et al. 2021; Güleç et al. 2022a). The critical challenges of climate change, environmental pollution and waste management are also some of the factors

responsible for the increase in the biomass based economy. The European Union's biomass based economy accounts for up to 4.2% of its yearly gross domestic production with generates a turnover of about €621 billion in value added benefits thereby creating more than 18 million jobs (Directorate-General 2018).

Biomass can be converted into green fuels and sustainable chemicals via thermochemical, biological or integrated conversion pathways. Biological processes such as fermentation and anaerobic digestion employ microorganisms for the degradation of biogenic materials into green fuels (Abdoulmoumine et al. 2015; Fan et al. 2016; Wang et al. 2017; Güleç et al. 2019a; 2019b; Sharma et al. 2019; Wilk et al. 2019; Antero et al. 2020; Arauzo et al. 2020; Cao et al. 2020; Shen 2020a). In contrast, thermochemical conversion pathways such as pyrolysis and gasification employ thermal and chemical energy at high temperatures for the decomposition of biomass. The integrated processes combine both thermochemical and biological pathways approach to minimise waste generation and promote energy and resource recovery (Okolie et al. 2022). A comprehensive review of different integrated pathways has been reported elsewhere (Okolie et al. 2022).

Although these technologies are promising, many challenges are hindering the commercialisation of biofuels and bioproducts obtained from these technologies. Some of the challenges include the heterogeneity and complex structure of biomass materials, cost effectiveness of the technologies compared to the business as usual pathways, challenges in the scalability of these technologies, and a limited and/or unstable supply of biofuels and bioproducts (Isikgor and Becer 2015; Guo and Song 2019; Güleç et al. 2021a, 2022c). Biomass heterogeneity, especially those obtained from different species or geographical locations, can result in significant variation in their characteristics; including biomass handling such as sizing, storage, feeding, and biochemical composition, which also affect the structure of the products produced during their conversion (Vassilev et al. 2015; Williams et al. 2015, 2016). The carbon budgets to meet climate targets are rapidly being exhausted (Lyngfelt 2020; Sigirci and Erdoğan 2022). With present emissions, the budget for limiting the global temperature increase to 1.5 °C will be used up by 2028 (Lyngfelt 2020). It is therefore highly unlikely to reach the climate targets on securing global net zero by mid-century and keeping 1.5 °C without significant negative greenhouse gas emissions technologies.

Since biomass-based power generation is defined as a carbon neutral process (Daioglou et al. 2019), carbon dioxide (CO_2) emissions from biomass and/or bio-based fuel combustion are excluded from any net CO_2 increase in the atmosphere (Lester et al. 2018; Sharma et al. 2020; Shen 2020b). Additionally, bioenergy with carbon capture and storage (BECCS) is a very attractive long-term carbon

negative technology for energy production. The crucial role of BECCS in the UK's net zero targets by 2050 has been recognised and reported by the UK climate change committee, the national grid's future energy scenario, and Inter-governmental Panel on Climate Change. The government's legally binding commitment to reduce emissions by 78% by 2035, in line with the climate change committee's 6th carbon budget advice, also reinforces the vital role of BECCS in delivering net zero economies in the UK. BECCS is a negative emissions technology (Fuss et al. 2018; Minx et al. 2018; Nemet et al. 2018), which generates power while permanently removing CO₂ from the atmosphere (Fajardy and Mac Dowell 2017).

The recent advances made in the field of BECCS cover advancements in biomass valorisation and optimisation, enhancement and integration of conventional biomass conversion technologies, and CO₂ capture, utilisation and storage. The advancement in biomass valorisation and optimisation, three distinct biomass feedstocks (whitewood, rapeseed residue, seaweed) were comprehensively investigated in three different thermal conversion technologies including hydrothermal conversion, pyrolysis, and torrefaction, for bioenergy production in integrated processes including BECCS (Güleç et al. 2022b, c, d, e, 2023a, b).

As for the enhancement and optimisation of conventional biomass processing technologies, gasification and pyrolysis technologies have been explored to enhance bioenergy production with the integration of solar energy to developed BECCS. Pinewood was pyrolysed using a solar simulator, which is calibrated by a high power xenon bulb as an energy source (Rony et al. 2019). Similarly, concentrated solar radiation were employed for pyrolysis of jatropha seeds (Chintala et al. 2017). The study explored the potential benefits of integrating biomass gasification into the future flexible power system, which includes BECCS or carbon capture and utilisation. The investigation focused on determining the profitability of integrating gasification into the electricity system and, if profitable, whether a flexible process design would be advantageous. Additionally, the study aimed to examine the potential impacts on the electricity system resulting from the implementation of these gasification processes. (Ahlström et al. 2022). As for the utilisation, the enhancement of gasification in oxyfuel BECCS cycles employing a direct recycling CO₂ utilisation process was investigated as a new method using computational models (Talekar et al. 2022). In addition to chemicals, the captured CO₂ is utilised to produce materials through mineralisation. Rosa et al (2022) introduce biogas concrete BECCS supply chains that permanently store biogenic CO₂ in recycled concrete aggregates through carbon dioxide mineralisation technologies (Rosa et al. 2022).

The advancement in CO₂ capture technologies focuses on exploring the advanced solvents and materials for CO₂

capture not only for BECCS application but also for net zero energy production. Ribeiro et al. (2022) reviewed the use of ionic liquids and biomass for the preparation of solid sorbents which can be used as CO₂ adsorbents and catalysts for conversion reactions of the same gas (Ribeiro et al. 2022). Lester et al. (2022) designed and operated a pilot scale CO₂ capture unit with advanced metal organic frameworks, "Pilot Scale Carbon Capture using Solid Sorbents" (Lester et al. 2022). The project aims to develop an advanced CO₂ capture technology for the application of power plants where biomass is combusted to produce energy and electricity. Thanks to the developments in the stable and commercial levels of metal organic frameworks synthesis and novel adsorption column design, the developed process provides relatively lower energy penalties than currently available and high technology readiness level amine scrubbing technology (Lester et al. 2022). Although the application of carbon capture and storage to biomass processing technologies is centred on conventional carbon capture and storage technologies such as post-combustion, pre-combustion and oxy-fuel combustion capture (Bhave et al. 2018; Finney et al. 2018; Kamer et al. 2018; Lea-Langton and Andrews 2018; Restrepo-Valencia and Walter 2021), the active gas separation steps in these technologies increase the energy penalty and inevitably leads to a decrease in overall plant efficiency (Romeo et al. 2008; Güleç et al. 2020a, b; Hosseini-Ardali et al. 2020; Chao et al. 2021). Chemical looping combustion on the other side potentially offers lower energy penalties (Fan et al. 2018; Schmitz and Linderholm 2018) and a lower CO₂ avoided cost (Bhave et al. 2018; Keller et al. 2019) compared to other conventional carbon capture and storage technologies. Therefore, over the last decade, there has been growing interest in chemical looping technologies, including chemical looping combustion of biomass and chemical looping gasification of biomass as attractive processes for biomass utilisation due to their low energy requirements.

In chemical looping combustion of biomass (Fig. 1a), biomass is converted to bioenergy using chemical looping combustion (also defined as advanced combustion) with simultaneous CO₂ capture, enabling green-carbon negative energy production. Chemical looping gasification of biomass (Fig. 1b) is a similar concept but applied to the gasification of biomass. In both processes, a solid oxygen carrier (commonly known as metal oxide) plays a vital role by providing the necessary oxygen for the reactions to occur. This crucial component enables efficient and controlled conversion of biomass into desired products, making it an essential element in these processes.

A comprehensive assessment of the costs of avoided CO₂ and the amount of captured CO₂ from eight different BECCS technologies (Fig. 2) showed that chemical looping combustion of biomass potentially has the lowest CO₂ avoided cost (about £62 per tonne CO₂) among the

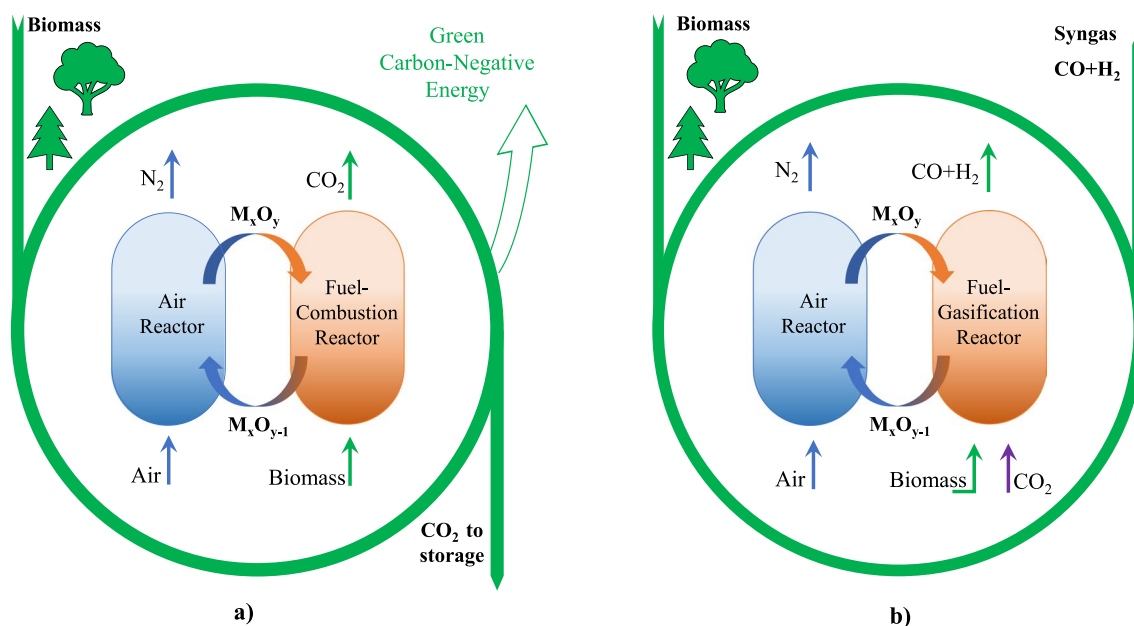


Fig. 1 General visualisation of **a** chemical looping combustion of biomass, in which biomass is converted to bioenergy using chemical looping combustion with simultaneous CO_2 capture, enabling green-carbon negative energy production. **b** Chemical looping gasification of biomass in which biomass is converted to syngas using chemical

looping technology. Abbreviations used in this figure; M_xO_y represents the oxidised oxygen carrier, M_xO_{y-1} represents the reduced oxygen carriers, which are carrying oxygen from air to the fuel reactor (combustion or gasification), carbon dioxide (CO_2), carbon monoxide (CO), hydrogen (H_2), nitrogen (N_2)

eight. In addition, chemical looping combustion of biomass currently has one of the largest relative uncertainty (± 33 per tonne CO_2) (Bhave et al. 2018). However, the cost of CO_2 captured with chemical looping combustion was determined as £145 per tonne CO_2 with an uncertainty of ± 66 per tonne CO_2 . Furthermore, with the application of chemical looping combustion, a potential cost reduction of the entire BECCS chain (17% less) was estimated over the benchmark circulating fluidised bed combustion plant with an amine scrubbing post-combustion process (Keller et al. 2019). These cost analyses showed that the chemical looping combustion of biomass is a relevant technology required to attain a net zero sustainable economy. A fluidised bed technology was employed as chemical looping gasification of biomass for pure hydrogen production with in situ CO_2 capture and sorbent regeneration (Acharya et al. 2009). The proposed technology has an energy efficiency of 87.5% with agricultural biomass feedstock. Chemical looping gasification of biomass can also be integrated with Fischer–Tropsch synthesis to produce a low-cost CO_2 free liquid fuel (Roshan Kumar et al. 2022). A dynamic model for syngas production from chemical looping gasification of biomass using Fe_2O_3 as an oxygen carrier was developed, and it demonstrated high energy and exergy efficiencies (Li et al. 2019). Despite the vast amount of information on different BECCS technologies, a

comparative review of the current status and technological progress is scantily reported.

The novelty of this manuscript lies in its comprehensive overview of the technology, status, and progress of biomass/biochar conversion through chemical looping combustion and chemical looping gasification for the purpose of BECCS. While the concept of chemical looping has been studied and applied in various industries, this manuscript specifically focuses on its application in the decarbonisation of bioenergy production. This manuscript stands out by providing a comprehensive overview of the technology, status, and progress of biomass/biochar conversion through chemical looping combustion and chemical looping gasification. It offers valuable insights into (1) biomass/biochar utilisation trends, (2) understanding of chemical looping technologies, and (3) the current state-of-the-art technologies along with future research directions.

1. *Evaluation of biomass utilisation trends:* The manuscript provides a systematic analysis of the research conducted on different biomass/biochar sources to utilise in chemical looping combustion and chemical looping gasification over the last decade.
2. *Comprehensive understanding of chemical looping combustion and gasification:* The manuscript presents a detailed explanation of the reaction mechanisms involved in chemical looping combustion and chemical

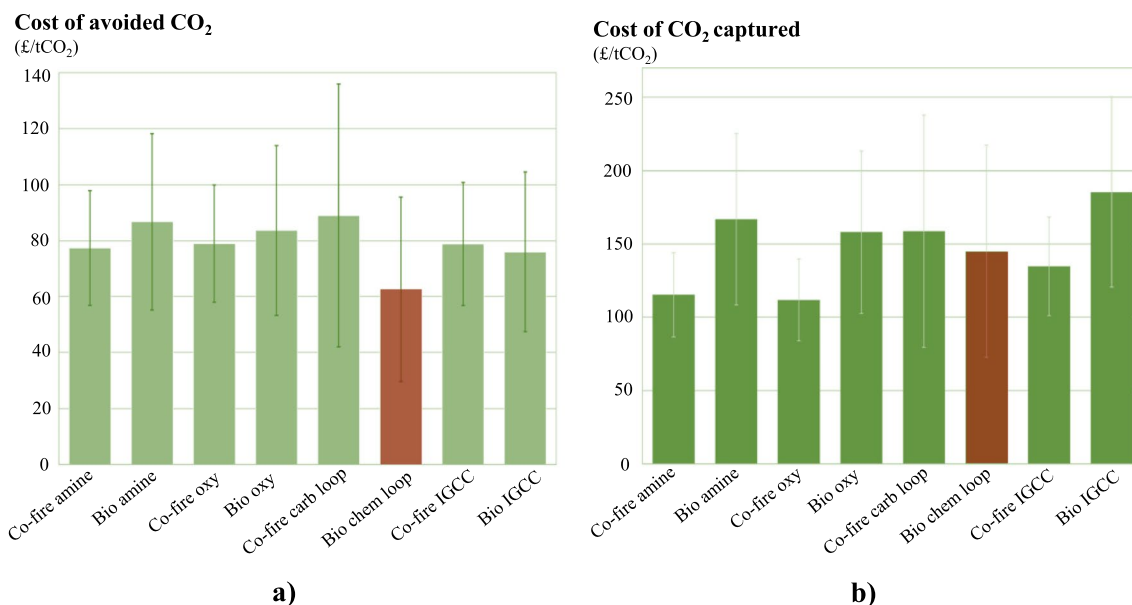


Fig. 2 **a** Cost of avoided CO₂ and **b** cost of CO₂ captured for the eight different bioenergy with carbon capture storage (BECCS) technologies, harmonised to 50 MWe (megawatt electric) scale, reprinted with permission of Elsevier from (Bhave et al. 2017). “Co-fire amine” represents biomass-coal co-firing combustion, with post-combustion amine scrubbing, “Bio-amine” represents biomass combustion with post-combustion amine scrubbing, “Co-fire oxy” represents biomass-coal co-firing oxy-combustion, with cryogenic O₂ separation, “Bio

oxy” represents biomass oxy-combustion, with cryogenic O₂ separation, “Co-fire carb loop” represents biomass-coal co-firing combustion, with post-combustion carbonate looping, “Bio chem loop” represents biomass chemical-looping-combustion using solid oxygen carriers, “Co-fire IGCC” represents biomass-coal co-firing integrated gasification combined cycle (IGCC), with physical absorption, “Bio IGCC” represents biomass integrated gasification combined cycle (IGCC), with physical absorption

looping gasification. It elucidates the underlying principles, advantages, and limitations of these chemical looping technologies.

3. *State-of-the-Art technologies and future research directions*: The manuscript goes beyond the basic overview and delves into the current state-of-the-art technologies in BECCS, specifically focusing on chemical looping gasification of biomass and chemical looping combustion of biomass. By addressing process challenges and discussing ongoing research efforts, it identifies potential areas for improvement and suggests future research directions.

Overview of chemical looping technologies

Chemical looping technologies have emerged as attractive processes for syngas production and clean energy applications through a wide range of process configurations. These technologies have been studied with various fuels such as methane, natural gas, syngas, coal, petroleum coke and waste biomass in various size pilot scale reactors (Mattisson and Lyngfelt 2001; Tian et al. 2008; Mattisson et al. 2009; Siriwardane et al. 2009; Wang et al. 2010, 2015; Chiu and Ku 2012; Güleç et al. 2020a, b). Chemical looping technologies that can be used for syngas production include

chemical looping gasification and chemical looping reforming. Moreover, chemical looping technologies for combustion purposes include chemical looping combustion, in situ gasification-chemical looping combustion, chemical looping oxygen uncoupling, and oxygen carrier aided combustion. The main characteristics of these technologies are summarised in Table 1.

There has been significant work on chemical looping technologies, with about 13,259 publications using the keywords “Chemical Looping” published according to the Web of Science between 2010 and 2022. Among these publications, 3575 publications were detected using the keyword “Chemical Looping Combustion” and 1353 publications were detected using the keyword “Chemical Looping Gasification”. Only 629 of chemical looping combustion publications are related to biomass “Biomass Chemical Looping Combustion”, and 604 of chemical looping gasification publications are related to biomass “Biomass Chemical Looping Gasification”. Among these publications, 183 chemical looping gasification of biomass and 248 chemical looping combustion of biomass research papers were published from countries in the European Union and European Economic Area.

Since BECCS aims to create a closed carbon cycle by capturing the CO₂ emitted during bioenergy production from biomass sources, chemical looping combustion and

Table 1 Main characteristics of chemical looping technologies

Chemical looping process and fuel type	Main characteristics and advantages
Chemical looping gasification (Solid fuels)	Partial oxidation by oxygen carriers instead of O ₂ . The process can also be adapted for pure N ₂ production and desired carbon monoxide/hydrogen (CO/H ₂) ratio Concentrated syngas (without N ₂ dilution nor pure O ₂ use) Auto thermal process Lower steam requirements Lower tar generation by reforming with oxygen carriers Lower CO ₂ emissions with respect to indirect gasification
Chemical looping combustion (Gas/Solid fuels)	Combustion of gas/solid fuels with oxygen carriers
Chemical looping reforming (Gas/Liquid fuels)	Partial oxidation of fuel with oxygen carriers The process can be adapted to produce a pure N ₂ stream and the desired CO/H ₂ ratio H ₂ production without CO ₂ emissions
In-situ gasification chemical looping combustion (Solid fuels)	Gasification of solid fuels in the fuel reactor to CO and H ₂ Combustion of gasification fuels and volatile matters in the fuel reactor with oxygen carriers
Chemical looping with oxygen uncoupling (Solid fuels)	Combustion of solid fuels with the gas phase oxygen release from oxygen carriers
Oxygen carrier aided combustion (Solid fuels)	Oxygen carrier redox reaction in different locations of the boiler, low cost oxygen carriers, tested at higher scale

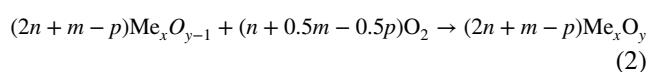
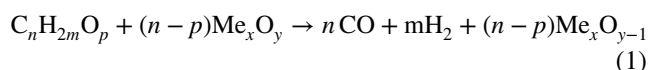
The chemical formulas used in the table; carbon dioxide (CO₂), carbon monoxide (CO), hydrogen (H₂), nitrogen (N₂), steam (H₂O), oxygen (O₂)

gasification play essential roles in BECCS technologies by facilitating CO₂ capture during biomass/biochar combustion and gasification processes. These chemical looping techniques offer efficient and flexible methods for biomass conversion while enabling the implementation of a closed carbon cycle, thus contributing to the development of a low-carbon and sustainable energy system. The combination of chemical looping combustion and chemical looping gasification in BECCS technologies offers several advantages: (1) carbon capture: chemical looping combustion and chemical looping gasification enable the capture of CO₂ during the combustion or gasification process. This captured CO₂ can be easily separated from other flue gases and stored, contributing to the reduction of greenhouse gas emissions. (2) Flexibility in biomass sources: chemical looping combustion and chemical looping gasification can be employed with a wide range of biomass sources, including agricultural residues, forest biomass, and energy crops. This versatility allows for the utilisation of various feedstocks in the BECCS process. (3) High Efficiency: The utilisation of chemical looping combustion and chemical looping gasification can lead to improved process efficiency in BECCS. By enabling the direct capture of CO₂ from the combustion or gasification process, the energy intensive separation processes typically associated with post-combustion capture can be minimised. (4) Waste Management: chemical looping combustion and chemical looping gasification provide sustainable waste management solutions by converting biomass into useful energy while capturing and storing CO₂. This promotes the

concept of a circular bioeconomy, where biomass resources are efficiently utilised and waste is minimised.

Chemical looping gasification of biomass

This section focuses on chemical looping gasification (Fig. 3), as it is used for syngas production via the use of solid fuels. The chemical looping gasification process is mainly based on oxygen transfer from an air reactor to a gasification reactor using solid metal oxides also called oxygen carriers, as oxidants. Firstly, the carbonaceous fuel either solid or gas is introduced to a fuel reactor and partially oxidised to carbon monoxide (CO) and hydrogen (H₂) as described in Reaction 1, by a metal oxide (Me_xO_y) (Wang et al. 2015; Mohamed et al. 2021). During this reaction, metal oxide (Me_xO_y) is reduced to a lower oxidation state (Me_xO_{y-1}). In a second reactor (air reactor), the reduced oxygen carrier (Me_xO_{y-1}) is oxidised by air or steam as provided in Reactions 2–3 (Wang et al. 2015; Mohamed et al. 2021). The reoxidised oxygen carrier (Me_xO_y) is ready for a new cycle between these two interconnected reactors in a solid circulatory loop, while the gas flows in these reactors are isolated using gas seals between the reactors.



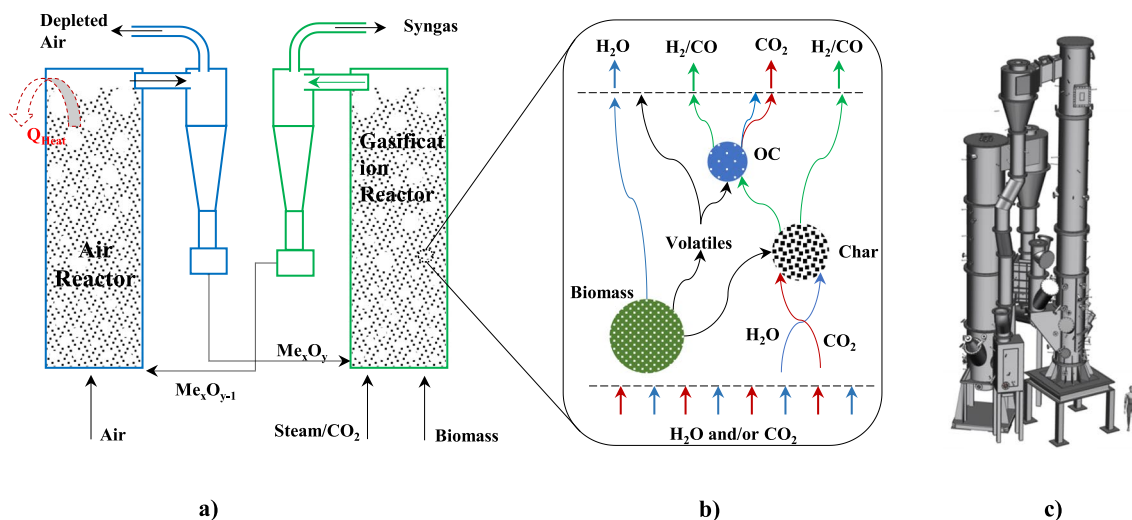
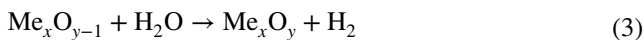


Fig. 3 **a** Schematic diagram of chemical looping gasification process, **b** potential reaction patterns in the fuel reactor (gasification reactor) between biomass, biochar, and oxygen carriers, and **c** a 1 MWth (megawatt thermal) pilot plant visualisation for chemical looping gasification of biogenic residues. “c” is reprinted with permission of MDPI from (Marx et al. 2021). In this figure; M_xO_y represents the

oxidised oxygen carrier, M_xO_{y-1} represents the reduced oxygen carriers, which are carrying oxygen from air to the fuel reactor (combustion or gasification). Q_{heat} represents the heat produced via metal oxide oxidation, OC represents oxygen carriers or metal oxides, carbon dioxide (CO_2), carbon monoxide (CO), hydrogen (H_2), nitrogen (N_2), steam (H_2O)



Here x and y represent the number of metal and oxygen atoms in the metal oxide. n , m , and p represent the number of carbon, hydrogen, and oxygen atoms in the hydrocarbon fuel. Me represents the metal atom in an oxygen carrier.

The unique oxygen separation system of chemical looping technologies makes them a preferred process with a smaller carbon footprint compared to the conventional gasification process. Pure oxygen supplied by the energy intensive air separation units in the conventional gasification process makes the process expensive. On the contrary, chemical looping gasification technology does not require a pure oxygen stream thanks to its unique separation property. The energy conversion efficiencies of the gasification process can be significantly increased through the application of chemical looping technologies. During chemical looping gasification, solid fuels such as biomass feedstocks and bio based chars can be directly introduced into the fuel reactor and physically mixed with the oxygen carriers for gasification. The required oxygen for the endothermic gasification reaction in the chemical looping gasification process is provided by the oxygen carriers circulating between two interconnected reactors, as shown in Fig. 3.

The biomass feedstocks are first thermally decomposed (devolatilisation and pyrolysis) to volatile matters mainly H_2 , CO, steam (H_2O), methane (CH_4), CO_2 , tar, and char in the fuel reactor (Reaction 4) (Nguyen et al. 2021; Samprón et al. 2021). As in the conventional gasification process, biomass

devolatilisation and pyrolysis, primary water gas, secondary water gas, Boudouard and CH_4 reforming reactions are endothermic, while the others are exothermic reactions. The gasification reactions specified in Table 2 are heterogenous reactions, except water gas shift and CH_4 reforming, which are homogenous reactions. Gibbs free energy (ΔG) of the primary and secondary water gas reactions (Reaction 5 and 6), Boudouard (Reaction 7), and methane reforming (Reaction 9) tend to decrease with an increase in the gasification temperature, where ΔG is below zero at a temperature higher than 700°C . As these three reactions are the main attractions for the syngas (CO/H_2) formation, it is recommended to keep the operating temperature of chemical looping gasification above 700°C . However, ΔG of the water gas shift (Reaction 8) and methane formation (Reaction 10) reactions tend to increase with the gasification temperature. Both reactions often lead to the formation of CO_2 and CH_4 .

Except for these five reactions, ΔG of the other reactions (in Tables 2 and 3) is always below zero, so these reactions are spontaneous during the gasification process. During chemical looping gasification potential reactions between pyrolysis products with Mn_2O_3 as an oxygen carrier could occur in addition to gasification reactions (Table 2). These reactions are presented in Table 3 (Reactions 11–18). Furthermore, chemical looping oxygen uncoupling could also release oxygen. The released oxygen can completely or partially oxidise the char or products (Reactions 19–23). The solid reduced oxygen carriers ($\text{Me}_x\text{O}_{y-1}$) are separated from reaction products in a cyclone, while the syngas (CO , H_2 , CH_4 , CO_2 , H_2O) exit the fuel reactor. The reduced oxygen

Table 2 Potential standard gasification reactions in the fuel reactor and enthalpy values

Name of reaction	Reactions	Enthalpy kJ/mol		References	Eq. number
		ΔH_{1273K}	ΔH_{298K}		
Devolatilisation and pyrolysis*	$\text{Biomass}_{(s)} \rightarrow \text{Volatiles}_{(g)} + \text{Char}_{(s)} + \text{H}_2\text{O}_{(g)}$	0 <	0 <	Nguyen et al. (2021), Samprón et al. (2021)	(4)
Primary (1°) water gas shift	$\text{C}_{(s)} + \text{H}_2\text{O}_{(g)} \leftrightarrow \text{H}_{2(g)} + \text{CO}_{(g)}$	135.5	131.3	Samprón et al. (2020), Marx et al. (2021)	(5)
Secondary (2°) water gas shift	$\text{C}_{(s)} + \text{H}_2\text{O}_{(g)} \leftrightarrow \text{H}_{2(g)} + \text{CO}_{2(g)}$	–	90	Wang et al. (2015)	(6)
Carbon (Boudouard) reaction	$\text{C}_{(s)} + \text{CO}_{2(g)} \leftrightarrow 2\text{CO}_{(g)}$	167.7	172.4	Wang et al. (2016a), Samprón et al. (2020)	(7)
Water gas shift	$\text{CO}_{(g)} + \text{H}_2\text{O}_{(g)} \leftrightarrow \text{H}_{2(g)} + \text{CO}_{2(g)}$	–32.2	–41.1	Samprón et al. (2020)	(8)
CH ₄ reforming	$\text{CH}_{4(g)} + \text{H}_2\text{O}_{(g)} \leftrightarrow 3\text{H}_{2(g)} + \text{CO}_{(g)}$	225.8	206.2	Samprón et al. (2020), Marx et al. (2021)	(9)
CH ₄ formation	$\text{C}_{(s)} + 2\text{H}_{2(g)} \leftrightarrow \text{CH}_{4(g)}$	–90.3	–74.8	Marx et al. (2021)	(10)

C, carbon

*Volatile matters represent mainly hydrogen (H₂), carbon monoxide (CO), moisture (H₂O), methane (CH₄), carbon dioxide (CO₂), and tar. ΔH represents enthalpy

Table 3 Potential reactions in fuel reactor of biomass gasification with Mn-based oxygen carriers at 1273 K

Name of reaction	Reactions	Enthalpy (ΔH , kJ/mol)	References	Eq. number
Reactions of pyrolysis and gasification products with Mn ₂ O ₃ in fuel reactor	$3\text{Mn}_2\text{O}_3(s) + \text{CO}_{(g)} \leftrightarrow 3\text{Mn}_3\text{O}_4(s) + \text{CO}_{2(g)}$	–192.2	Wang et al. (2016a)	(11)
	$\text{Mn}_2\text{O}_3(s) + \text{CO}_{(g)} \leftrightarrow 2\text{MnO}_{(s)} + \text{CO}_{2(g)}$	–101.9		(12)
	$3\text{Mn}_2\text{O}_3(s) + \text{H}_{2(g)} \leftrightarrow 2\text{Mn}_3\text{O}_4(s) + \text{H}_2\text{O}_{(g)}$	–160.0		(13)
	$\text{Mn}_2\text{O}_3(s) + \text{H}_{2(g)} \leftrightarrow 2\text{MnO}_{(s)} + \text{H}_2\text{O}_{(g)}$	–69.7		(14)
	$3\text{Mn}_2\text{O}_3(s) + \text{CH}_{4(g)} \leftrightarrow 2\text{Mn}_3\text{O}_4(s) + \text{CO}_{(g)} + 2\text{H}_{2(g)}$	65.8		(15)
	$4\text{Mn}_2\text{O}_3(s) + \text{CH}_{4(g)} \leftrightarrow 8\text{MnO}_{(s)} + \text{CO}_{2(g)} + 2\text{H}_2\text{O}_{(g)}$	–85.2		(16)
	$3\text{Mn}_2\text{O}_3(s) + \text{C}_{(s)} \leftrightarrow 2\text{Mn}_3\text{O}_4(s) + \text{CO}_{(g)}$	–24.4		(17)
	$2\text{Mn}_2\text{O}_3(s) + \text{C}_{(s)} \leftrightarrow 4\text{MnO}_{(s)} + \text{CO}_{2(g)}$	–36.1		(18)
Oxidation in fuel reactor (if metal oxide release O ₂)	$\text{C}_{(s)} + \text{O}_{2(g)} \leftrightarrow \text{CO}_{2(g)}$	–395.2	Condori et al. (2021), Golgiyaz et al. (2023a)	(19)
	$\text{C}_{(s)} + 0.5\text{O}_{2(g)} \rightarrow \text{CO}_{(g)}$	–111		(20)
	$\text{CO}_{(g)} + 0.5\text{O}_{2(g)} \rightarrow \text{CO}_{2(g)}$	–284		(21)
	$\text{CH}_{4(g)} + 2\text{O}_{2(g)} \leftrightarrow \text{CO}_{2(g)} + \text{H}_2\text{O}_{(g)}$	–803		(22)
	$\text{H}_{2(g)} + 0.5\text{O}_{2(g)} \rightarrow \text{H}_2\text{O}_{(g)}$	–242		(23)
Re-oxidation	$4\text{Mn}_3\text{O}_4(s) + \text{O}_{2(g)} \rightarrow 6\text{Mn}_2\text{O}_3(s)$	–192.7* (–201.8 ⁺)	Fritsch and Navrotsky (1996)	(24)
	$6\text{MnO}_{(s)} + \text{O}_{2(g)} \rightarrow 2\text{Mn}_3\text{O}_4(s)$	–432.9* (–441.4 ⁺)		(25)

C, carbon; Mn₂O₃, Manganese(III) oxide; CO, carbon monoxide; Mn₃O₄, Manganese(II, III) oxide; MnO, Manganese(II) oxide; H₂, hydrogen; CH₄, Methane; H₂O, steam; CO₂, carbon dioxide; (O₂, oxygen

*Enthalpy (ΔH , kJ/mol of oxygen) values at 977 K. ⁺Enthalpy (ΔH , kJ/mol of oxygen) values at 298 K

carriers are then transferred to the air reactor in which the reduced oxygen carriers are oxidised with air (an exothermic reaction, presented in Reactions 24–25). Following the oxidising, the reoxidised oxygen carriers are circulated back to the fuel reactor, where the loop is closed in terms of the oxygen carrier.

Transferring unconverted char from the fuel reactor to the air reactor can be eliminated by a carbon stripper located between the cyclone attached to the fuel reactor and air reactor, which keeps the air reactor clean from fuel components.

In the chemical looping gasification, most of the process heat is extracted inside the air reactor and some from the off gas while the extracted heat can be utilised for energy (power/steam) production. The chemical looping gasification process could be operated either under pressurised conditions or atmospheric pressure, which avoids the compression of the syngas or the compression of air to the air reactor, respectively. A summary of selected biomass feedstocks in the chemical looping gasification is presented in Table 4.

The works existing in the literature on chemical looping gasification operation in continuous units usually analyse the effect of the operating conditions on syngas composition and gasification parameters. The main operating conditions of a continuous chemical looping gasification of biomass unit are defined as the oxygen-to-biomass ratio, steam-to-biomass ratio, different oxygen carriers and gasification temperature. The impact of these operating conditions on syngas quality and composition has been reported by several researchers (Sampron et al. 2020). Among these operating conditions, the oxygen-to-biomass ratio is one of the most prominent parameters and could be justified by (1) biomass feeding rate (Huseyin et al. 2014; Wei et al. 2015a, b; Shen et al. 2017) and (2) circulating rates of oxygen carriers (Huijun et al. 2015; Ge et al. 2016) in the chemical looping gasification of biomass. In the first one, the ratio of oxygen to biomass can be adjusted by changing the biomass feed rate, keeping the oxygen carrier circulation rate constant, which could also vary the steam-to-biomass ratio (Huseyin et al. 2014; Wei et al. 2015a, b; Shen et al. 2017). In the second one, the ratio of oxygen to biomass can be adjusted by changing the oxygen carrier circulating rate, keeping the biomass feeding rate constant. The oxygen carriers could be modified by inert supports (i.e. SiO_2) not to increase the solid inventory in the fuel reactor of chemical looping gasification (Huijun et al. 2015; Ge et al. 2016; Boztepe et al. 2021).

A total of 55 h of the continuous run was investigated in a 1.5 kWth chemical looping gasification unit by some researchers (Condori et al. 2021), and the impact of process conditions on the biomass chemical looping gasification was also evaluated (Sampron et al. 2020). In the chemical looping gasification experiments, relatively high biomass conversion was reached at about 89.5–99.8% at the temperatures of 820–940 °C with a steam-to-biomass ratio of 0.6–0.9 (Condori et al. 2021). Furthermore, relatively high gasification efficiency and carbon conversion efficiency are determined as 82.6 and 87.6% in the direct chemical looping gasification of sawdust of pine with natural hematite (Huang et al. 2013a, b, c). Though, the increase in the gasification temperature did not provide a significant impact on the fuel conversion (which is stable at over 88%). Also elevating temperature enhances the carbon conversion efficiency to a maximum of 100% at 940 °C when pinewood was the precursor (Sampron et al. 2020; Condori et al. 2021). The best operating temperature was proposed as 900 °C for both wheat straw and pine forest residue (Di Giuliano et al. 2021). Furthermore, higher carbon conversion (85%) was observed with optimum superficial velocity (Nguyen et al. 2021). Relatively high carbon conversion efficiencies (above 95%) and fuel conversion (nearly 100%) were also observed as a function of the oxygen-to-biomass ratio of 0.58 when pinewood biomass was the feedstock (Sampron et al. 2020). In another study, the carbon conversion was determined as 80–82% for wheat

straw and 63–71% for pine forest residue (Di Giuliano et al. 2021) during chemical looping gasification. The carbon conversion decreased from 87.7 to 77.2% in 20 cycles due to the attrition and structural changes of the oxygen carrier (Huang et al. 2013a, b, c).

The increase in the oxygen-to-biomass ratio increased the carbon conversion while reducing the H_2 production (Nguyen et al. 2021). Similarly, the increase in the oxygen-to-biomass ratio from 0.21 to 0.58 decreases the syngas yield from 0.94 to 0.45 Nm^3/kg , cold gas efficiency from 79.1 to 44.4% (Sampron et al. 2020), and CO and H_2 concentrations (Condori et al. 2021). On the other hand, another study showed that the presence of steam can improve carbon conversion from 35 to 64% and gas yield from 0.62 to 2.46 g/L under mixed oxygen source and steam conditions (Huang et al. 2016). The increase in the steam-to-biomass ratio enhances the H_2 and CO_2 concentrations with decreasing CO concentration and produced a higher H_2/CO ratio from 1.0 to 1.5 (Condori et al. 2021). The increase in the steam concentration increases the gas yield (15.9%) and H_2 content, 43.0% for the iron ore and 43.9% for the ilmenite at a steam-to-biomass ratio of 1.4 (Nguyen et al. 2021). Furthermore, some researchers showed that elevating steam concentration would improve the gasification rate resulting in the enhancement of the fuel conversion efficiency from 75.7 to 88% (Sampron et al. 2020).

Relative concentrations of syngas H_2 (22.0%), CH_4 (12.3%), and CO (50.6%) were produced by the chemical looping gasification of sawdust pine with natural hematite (Huang et al. 2013a, b, c). The chemical looping gasification of Pinewood with ilmenite provides gas products composed of 37–40% CO_2 ; 27–30% H_2 ; 17–21% CO; 10–12% CH_4 ; and 2–3% $\text{C}_2\text{--C}_3$ with a maximum syngas yield of 0.89 Nm^3/kg dry biomass (Condori et al. 2021). However, syngas having higher H_2 and lower CO_2 (37% of H_2 , 34% of CO_2 , 21% of CO, and 7% of CH_4) were observed in the chemical looping gasification of Pinewood with synthetic Fe-based oxygen carriers, leading to a syngas yield of 0.8 Nm^3/kg dry biomass and cold gas efficiency of 68% (Sampron et al. 2020). Similarly, the H_2/CO molar ratio was 0.9–1.0 for wheat straw and pine forest residue and the highest H_2 yield was determined as 40% at 900 °C for the biomass feedstocks (Di Giuliano et al. 2021).

Techno-economic analysis of chemical looping gasification has also been studied either as a standalone process or integrated with other biomass conversion processes in an integrated circular economy (Roshan Kumar et al. 2022). Sarafraz and Christo (Sarafraz and Christo 2020) developed a chemical looping gasification utilising indium liquid slurry. Liquid metals have the advantages of easy oxidation and reduction without disrupting their structure with impurities addition and separation. Furthermore, they provide a decent thermal conductivity and promising capacity

Table 4 Chemical looping gasification of various biomasses

Type of biomass	Types of reactor	Operating conditions	Oxygen carrier	Research outcomes and remarks	References
Pinewood	Bubbling fluidised bed (Pilot scale—1.5 kWth)	Steam 820–940 °C	Ilmenite	Biomass conversion was about 89.5–99.8% at 820–940 °C with an SBR of 0.6–0.9. Higher temperature enables full conversion. 100% of conversion at 940 °C with 0.6SBR and 0.34OBR. The gas products are composed of 37–40% CO ₂ ; 27–30% H ₂ ; 17–21% CO; 10–12% CH ₄ ; and 2–3% C ₂ –C ₃ with a max synthesis yield of 0.89 Nm ³ /kg dry biomass	Condori et al. (2021)
		940 °C OBR ^c (0.15–0.48) SBR ^b (0.05–0.9)			
Wheat Straw Pine Forest Res	Bubbling fluidised bed (Lab scale)	Nitrogen, Steam, Air 700–900 °C	Ilmenite Sibelco LD-slag Sand	900 °C was recommended as the best operating temperature for both Wheat Straw and Pine Forest Residue. 80–82% and 63–71% of carbon conversion for Wheat Straw and Pine Forest Residue, respectively. Gas yield: 0.038–0.042 mol/g of Wheat Straw and 0.036–0.042 ml/g of Pine Forest Residue with the oxygen carriers. The ratio of H ₂ /CO was 0.9–1.0 for Wheat Straw and Pine Forest Residue	Di Giuliano et al. (2021)
		700–900 °C			
Torrefied woodchips	Bubbling fluidised bed (Pilot scale)	Steam 850 °C	Iron ore, Ilmenite	Carbon conversion was 85% and higher OBR tends to reduce H ₂ production. Syngas fuels are H ₂ > CO > CH ₄ with iron-based oxygen carriers. Higher SBR enables a gas yield of 15.9% and H ₂ of 42.89% for the iron ore and 43.87% for the ilmenite	Nguyen et al. (2021)
		OcBR ^a (0–6) SBR ^b (0.9–1.4)			
Sawdust of pine	Fluidised bed	Steam/N ₂ 840 °C	Natural hematite	Gasification efficiency: 82.6% and carbon conversion efficiency: 87.6%. Carbon conversion decreased from 87.6% to 77.2% in 20 cycles. The concentration of syngas was CO > H ₂ > CH ₄ : 50.6 > 22.0 > 12.3, respectively	Huang et al. (2013a, b), c)
		840 °C			
Rice straw	Fixed bed	Steam 600–900 °C	Fe ₂ O ₃ -CaO	Hydrogen yield (23.1 mmol/g biomass) under the optimum ratio of Fe/Ca (1:1 of Ca ₂ Fe ₂ O ₅). H ₂ production decreased due to the low stability of Fe-Ca	Hu et al. (2020)

Table 4 (continued)

Type of biomass	Types of reactor	Operating conditions	Oxygen carrier	Research outcomes and remarks	References
Pinewood	Fluidised bed	Simulation-based on auto-thermal conditions	Fe-based synthetic	The syngas yield is higher in the process where the oxygen feed in the air reactor is controlled through the air flow (OCM-1) compared to controlling the oxygen supplied to the fuel reactor by oxygen carriers (OCM-2). Syngas yield increases with the increase in the oxygen carrier circulation between fuel-air reactors in the OCM-1 configuration. Cold gas efficiency (79.8–86.2) and fuel value (0.33–0.38)	Samprón et al. (2021)
Pinewood	Bubbling fluidised bed (Pilot scale – 1.5kWth)	Steam 820–940 °C OcBR ^a (0.2–0.6) SBR ^b (0.05–0.65)	Fe-based synthetic (Fe ₂ OAl ₂ –20 wt% Fe ₂ O ₃)	Carbon conversion efficiency (above 95%) and fuel conversion (nearly 100%). A syngas composed of 37% H ₂ , 21% CO, 34% CO ₂ and 7% CH ₄ , and tars below 2 g/Nm ³ . Syngas yield of 0.8 Nm ³ /kg dry biomass and cold gas efficiency of 68%	Samprón et al. (2020)
Biochar	Fixed bed	Steam/N ₂ 850 °C	Iron ore	The optimised iron ore oxygen excess number value was found as 0.71, where the synthesis gas volume: 153 mL, and the relative content of H ₂ (20.0%) and CO (41.9%). The presence of steam can improve the carbon conversion (from 35 to 64%) and gas yield increases (from 0.62 g/L to 2.46 g/L)	Huang et al. (2016)
Wood Pellet Pine Forest Res Wheat Straw	Fluidised bed reactor (Pilot scale – 1.0MWth)	Steam 950 °C	Ilmenite	Autothermal CLG demonstrates a low range of oxygen carrier to fuel equivalence ratio, steam-to-biomass feed ratio, and operation temperatures compared to externally heated lab-scale units. CLG process can be operated by auto thermal conditions controlling oxygen carrier to fuel equivalence ratio, thermal load, and solid circulation. It is possible to reach high cold gas efficiency and syngas quality by increasing the gasification temperature, however, this results in high relative heat losses in the IMWth scale	Marx et al. (2021)

Table 4 (continued)

Type of biomass	Types of reactor	Operating conditions	Oxygen carrier	Research outcomes and remarks	References
Wood Pellet Wood Char	Batch fluidised bed TGA	50% Steam in Nitro- gen 820– 970 °C	Ilmenite LD-slag Sand Olivine	LD slag consists of Ca, Mg, Al and Mn besides iron. The presence of CaO makes LD slag a promising oxygen carrier candidate for biomass CLG to achieve syngas with a high H ₂ /CO ratio. Compare to sand and olivine, LD slag provides a high char conversion rate (3.5 g/g.s) at 970 °C. The highest ratio of H ₂ /CO is 28 at 820 °C. The highest CO/C ratio is 0.45 at 970 °C. The high ratio of H ₂ /CO by LD slag at low temperature (820 °C)	Hildor et al. (2020)

CLG, chemical looping gasification; CaO, calcium oxide; TGA, thermogravimetric analyser; kWth, Kilowatt thermal; MWth, Megawatt thermal; LD-slag, steel slag as waste through the blast furnace and Linz–Donawitz (LD) process; H₂, Hydrogen; CO, carbon monoxide; H₂O, moisture; CH₄, methane; CO₂, carbon dioxide

^aOcBR, Oxygen carrier-to-biomass ratio; ^bSBR, Steam-to-biomass ratio; ^cOBR, Oxygen-to-biomass ratio

for cyclic reduction and oxidation reactions. The proposed design was able to produce syngas and electricity simultaneously. The peak performance of the proposed system is attained when the fuel and air reactors are operated at near isothermal conditions. Economic evaluation of the proposed design demonstrated that the levelised cost of electricity is comparable to similar energy systems. The syngas produced via chemical looping gasification has a higher calorific value than that generated by a conventional air gasification derived syngas (Aghabararnejad et al. 2015). Although the total capital investment of the chemical looping gasification was higher than conventional air gasification, the yearly operating cost of chemical looping gasification was lower which repays the difference in total capital cost in less than six years.

Chemical looping combustion of biomass

The chemical looping combustion concept was first described by Richter and Knoche (Richter and Knoche 1983), which is based on oxygen transfer from the air reactor to the fuel reactor via a solid oxygen carrier. In this process, air does not need to mix with fuel for combustion. In the chemical looping combustion of biomass (Fig. 4), oxygen transfers from the air reactor to the fuel reactor via a solid oxygen carrier. The fuel such as biomass or biochar is first introduced to the fuel reactor and oxidised to CO₂ and H₂O (Reaction 26) by a metal oxide (Me_xO_y) (Wang et al. 2015), while the oxygen carrier is reduced to Me_xO_{y-1}. After a condensation and purification step, the pure CO₂ is ready for transport and storage (Adanez et al. 2012). Subsequently, in the air reactor, the reduced oxygen carrier (Me_xO_{y-1}) is reoxidised by oxygen in the air stream (Reaction 27). The reoxidised oxygen carrier (Me_xO_y) gets ready for a new combustion cycle. The total reaction occurring in the chemical looping combustion of biomass process (Reaction 28) is thermodynamically equal to the combustion of biomass with oxygen.

In essence, chemical looping combustion of biomass is a process of indirect combustion, in which air and fuel are never mixed, (Pröll et al. 2009); thus, the technique does not need an additional separation process (Mattisson et al. 2009; Ksepko et al. 2012) as the chemical looping combustion method may allow for natural separation of CO₂ and H₂O from other non-condensable gases; N₂ and excess O₂ (Mattisson et al. 2009). In contrast, other carbon capture and storage technologies require CO₂ separation technologies, i.e. post-combustion like amine scrubbers and pre-combustion or oxy-combustion like air separation unit, which are a significant characteristics increasing the energy penalty and CO₂ capture avoided costs (Mattisson et al. 2009; Pröll et al. 2009; Ksepko et al. 2012).

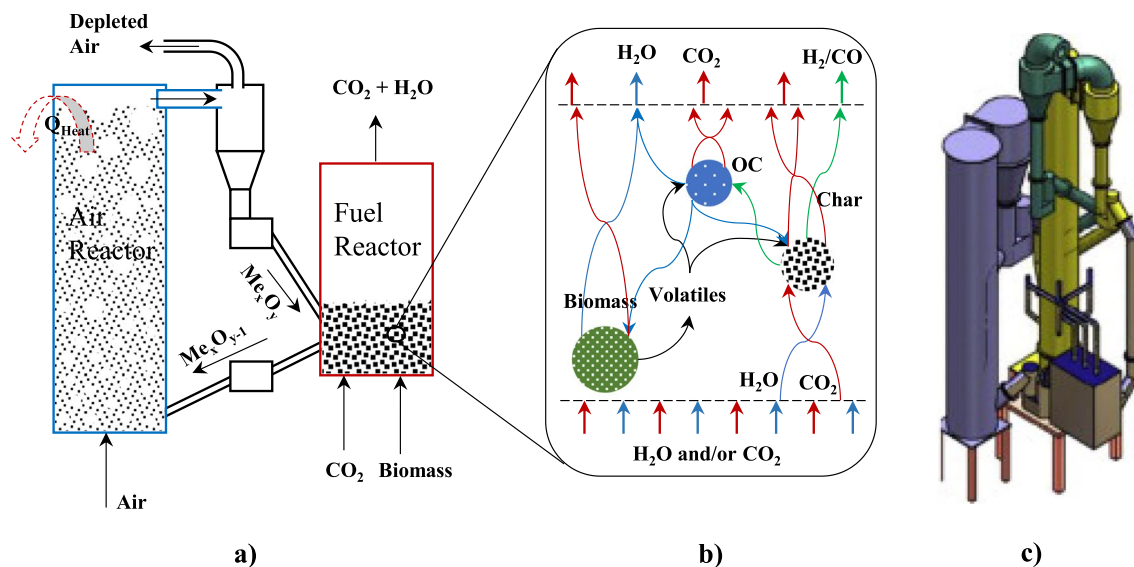
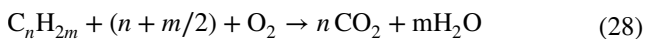
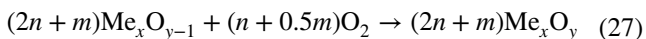
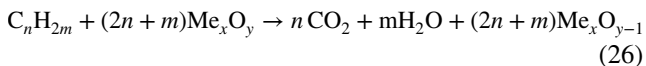


Fig. 4 **a** Schematic diagram of the chemical looping combustion process, **b** potential combustion reactions between biomass, biochar, and oxygen carriers in the fuel reactor, and **c** a 1.0 MWth (megawatt thermal) pilot scale chemical looping combustion unit visualisation for solid fuels. **c** Reprinted with permission of Elsevier from (Adanez et al. 2012). In this figure, M_xO_y represents the oxidised oxygen carrier,

M_xO_{y-1} represents the reduced oxygen carriers, which are carrying oxygen from air to the fuel reactor (combustion), Q_{heat} represents the heat produced via metal oxide oxidation, OC represents oxygen carriers or metal oxides, carbon dioxide (CO_2), carbon monoxide (CO), hydrogen (H_2), nitrogen (N_2), steam (H_2O)



Here x and y represent the number of metal and oxygen atoms in the metal oxide. n and m represent the number of carbon and hydrogen atoms in the hydrocarbon fuel. Me represents the metal atom in an oxygen carrier.

The chemical looping combustion studies focus on the development of oxygen carriers, reactor design, and application of chemical looping combustion for fuels (solid, liquid, and gas). A plethora of research studies stated that chemical looping combustion is a promising technology to generate a high concentration of CO_2 in combustion and hydrogen production processes (Rydén and Lyngfelt 2006; Chiu and Ku 2012; Bhavsar et al. 2014), especially when a suitable metal oxide is used as an oxygen carrier (Table 5) (Pröll et al. 2009). As a result of these studies, the chemical looping combustion process, where metal oxides can be used as an oxygen supplier instead of air to combust the fuel stream (Ksepko et al. 2012), has been proposed as a potential alternative process to decrease CO_2 capture costs (Kerr 2005) and energy penalties. In order to demonstrate the industrial feasibility of chemical looping combustion from an

economic and technical perspective, suitable oxygen carriers are critical issues for specific fuels and reactor systems.

Oxygen carriers

Oxygen carriers usually made from metal oxides; either single or combined forms of metal oxides, such as Fe_2O_3 , CuO , NiO , Mn_2O_3 , Co_3O_4 and inert binders such as Al_2O_3 , MgAl_2O_4 , SiO_2 , ZrO_2 (Adánez et al. 2004, 2009; Ksepko et al. 2012; Wang et al. 2015). Moreover, natural minerals such as iron ore, manganese ore, and copper ore can be used as oxygen carriers (Mendiara et al. 2012; Tian et al. 2013; Wang et al. 2016b). The oxygen carriers should accomplish the following characteristics to be feasible for industrial applications of chemical looping technologies (Cho et al. 2004; Adánez et al. 2006; Adanez et al. 2012; Wang et al. 2015); sufficient oxygen transport capacity for gasification, high redox activity, resistance to fragmentation and attrition, partial fuel conversion to CO and H_2 for chemical looping gasification and complete fuel conversion to CO_2 and H_2O for chemical looping combustion, negligible carbon deposition, a low tendency for agglomeration, easy preparation method and low cost.

Iron (Fe-) based oxygen carriers mostly have a lower cost, lower environmental impact, better mechanical strength, and higher melting point compared to other oxygen carriers (Adánez et al. 2004; Cho et al. 2004; Ksepko et al. 2012;

Table 5 Chemical looping combustion of various biomasses

Type of biomass	Types of reactor	Operating conditions	Oxygen carrier	Research Outcomes and Remarks	References
Pine sawdust, Olive stones, Almond shells	Fluidised bed (1.5 kWth CLC)	Combustion temperature (775–850 °C) Oxidation temperature (950 °C) Oxygen carrier to fuel ratio (2.3–7.6) Operating time 40 h of combustion	$\text{Cu}_{1.5}\text{Mn}_{1.5}\text{O}_4\text{Mn}_3\text{O}_4$ (Spray granulation)	Fuel reactor outlet did not provide any unburnt gasses even at the lowest temperature of 775 °C, which results in relatively high CO_2 capture and complete combustion efficiencies (100%) During the CLC of pine sawdust, CO_2 capture efficiency was > 95% in most of the cases at operating temperatures The developed oxygen carrier could generate enough oxygen to release through CLOU properties and excess into the fuel reactor, which can fully convert the biomass fuel. Furthermore, 100% combustion was also observed for olive stones and almond shells with a CO_2 capture efficiency of > 90% at 850 °C	Adánez-Rubio et al. (2018)
Wood pellets, Wood char, Straw pellets	Circulating fluidised bed (60 kW CLC pilot)	Fuel feed rates (1.9–3.7 g/s) Residence time (39 to 179 min) Process temperature (750–850 °C)	Ilmenite and Braunitz (Natural Ore)	Alkali emissions were investigated. In the conversion of wood pellets with ilmenite, no alkalis were detected However, wood pellets combustion with braunitz shows that the sample contained KCl, NaCl, KOH, and NaOH The samples only contained KOH and KCl for the CLC of wood char with ilmenite. As for straw pellets, almost all of the alkalis detected occur as KCl	Gogolev et al. (2022a)

Table 5 (continued)

Type of biomass	Types of reactor	Operating conditions	Oxygen carrier	Research Outcomes and Remarks	References
Olive stones, Almond shells, Pine sawdust	Fluidised bed (0.5 kWth CLC)	Combustion temperature (900–980 °C) Oxidation temperature (800 °C) Oxygen carrier to fuel ratio (1.0–1.3) Operating time 65 h (40 h of combustion)	Fe ore Tierra (calcined 950 °C)	Combustion behaviour of these three biomass feedstocks did not show any differences The outlet of the fuel reactor presents 70% of CO ₂ in addition to unburnt compounds: H ₂ , CO and CH ₄ Almost 100% of CO ₂ capture efficiency was achieved in the fuel reactor by increasing the temperature above 950 °C High volatile matter in biomass feedstocks required higher oxygen demand 25–30%	Mendiara et al. (2018)
Swedish wood char, German wood char, Steam-cured black pellets	Dual fluidised bed continuous unit (10 kWth)	Combustion temperature (820–975 °C) Operating time 42 h (21 h of combustion)	Manganese-based ores and ilmenite, (calcined at 950 °C, wet sieving)	Manganese ore shows higher reactivity than the often used ilmenite (Fe-based ores, which decreases the oxygen demand by 8–10% compared to ilmenite) Manganese ore used in this study showed a longer lifetime, i.e. 370–830 h compare to other Mn-based ores Biochars demand lower oxygen (2.6%) while providing relatively high carbon capture 99% Manganese ore did not show defluoridation even after 42 h of hot chemical looping operation	Mei et al. (2021)

Table 5 (continued)

Type of biomass	Types of reactor	Operating conditions	Oxygen carrier	Research Outcomes and Remarks	References
Pinewood, Olive stones, Almond shells	Fluidised bed (0.5 kWth CLC)	Combustion temperature (850–950 °C) Oxidation temperature (800 °C) Oxygen carrier to fuel ratio (1.0–3.5)	Manganese-based ores (calcined 800 °C)	CO ₂ capture efficiency reached about 100% for pine sawdust and almond shells combustion with Mn-ores The CLC requires excess oxygen demand of 10–20% In order to reduce the oxygen demand, the outlet of the fuel reactor was operated as recycling and the results show a 30% reduction in the total oxygen demand value No NOx emissions and low tar content were observed in the outlet stream of the fuel reactor	Pérez-Astray et al. (2020)
Pine sawdust, Rice husk	Dual circulating fluidised beds	Combustion temperature (840–950 °C) Feeding rate (60–180 g/h)	Natural hematite (calcined 950 °C)	92.5% of carbon conversion and 98.5% of carbon capture were achieved at 900 °C The combustion temperature above 880 °C is optional in this study thanks to the unique multistage fluidised bed reactor design, which improved solid gas interactions. In addition to biomass conversion, there was no sintering observed on the natural hematite oxygen carrier after operating in chemical looping	Jiang et al. (2018)

Table 5 (continued)

Type of biomass	Types of reactor	Operating conditions	Oxygen carrier	Research Outcomes and Remarks	References
Wood chips	Circulating and batch fluidised bed (12 MWth)	Combustion temperature (850–860 °C) Total bed inventory (2000 kg of LD slag) Batch Unit: Oxidising (5% O ₂ , 95% N ₂ , 800–950 °C) Inert (100% N ₂ , 800–950 °C)	LD-slag (Steel converter slag: calcium and iron based)	The LD-slag can be used as an oxygen carrier, which shows a reduction of magnetite to wüstite. It provides low attrition and low agglomeration. However, some of the inorganic materials in biomass ash (i.e. potassium, sulphur and phosphorus) were accumulated over the oxygen carrier. The activity decreased with the operation. Although elemental sulphur addition enhanced the reactivity after 60 h of operation, it was still lower than the fresh one.	Hildor et al. (2019), Qasim et al. (2021)
Black pellet, Pine forest residue, Straw pellets	Dual circulating fluidised beds (10 kWth)	Combustion temperature (870–920–970 °C)	Linz–Donawitz slag (calcined 950 °C, dry-sieved)	Alkali release from chemical looping combustion of biomass and chemical looping gasification of biomass were investigated. Higher temperature results in higher alkali release in the gas phase from the fuel reactor, which was attributed to KCl evaporation and alkali salt decomposition. Similar gas phase alkali emissions were observed from CLC and CLG due to similar char conversion.	Gogolev et al. (2022b)
Commercial biochar (hardwood)	Fluidised bed	Combustion temperature (750–895 °C) Fluidisation gas (~ 1.5 vol% O ₂ in N ₂)	CuO/mayenite, SrFeO ₃ - δ , Fe ₂ O ₃ (prepared from pyrite ore based)	The required burn-out time followed as CuO < SrFeO ₃ - δ < Fe ₂ O ₃ \approx silica sand. The better performance of CuO was attributed to the higher oxygen releasing capacity of CuO via CLOU. CO produced by incomplete combustion was reacted with Fe ₂ O ₃ .	Kwong et al. (2022)

CLC, chemical looping combustion; CLOU, chemical looping oxygen uncoupling; CLG, represents chemical looping gasification; kWth, Kilowatt thermal; MWth, Megawatt thermal; LD-slag, steel slag as waste through the blast furnace; LD, Linz–Donawitz process; H₂, Hydrogen; CO, carbon monoxide; H₂O, moisture; CH₄, methane; CO₂, carbon dioxide; O₂, oxygen; Fe₂O₃, iron(III) oxide; CuO, copper(II) oxide; KCl, potassium chloride; NaCl, sodium chloride; NaOH, sodium hydroxide; KOH, potassium hydroxide

Wang et al. 2016b). The reactivity of Fe-based oxygen carriers is enough at atmospheric and pressurised conditions (Cho et al. 2004; Scott et al. 2006; Wang et al. 2016b). The Fe-based oxygen carriers react rapidly with intermediate gasification products; CO and H₂ (Leion et al. 2007). Additionally, the combustion of coal with Fe₂O₃ may be improved when the coal is blended with biomaterials such as straws (Wang et al. 2014). Furthermore, the alkali carbonate such as K₂CO₃, Li₂CO₃, and Na₂CO₃, treatment on Fe₂O₃ oxygen carriers can enhance the combustion rate of char (Yu et al. 2012). In addition to synthetic Fe-based oxygen carriers, some natural Fe-based oxygen carriers were also investigated as oxygen carriers. Both hematite and Fe-based synthetic oxygen carriers showed high combustion efficiency with the lowest amount of oxygen carrier in the reactor bed (Mendiara et al. 2012). As for the carbon formation, while there is no or minimal carbon deposition on Fe-based synthetic oxygen carriers (Cho et al. 2005; Brown et al. 2010; Wang et al. 2016b), carbon formation of natural Fe-based oxygen carriers is associated with reduction time (Tian et al. 2013). Most studies on chemical looping technologies have suggested that Fe-based oxygen carriers remain durable through multiple reduction and oxidation cycles (Rubel et al. 2009; Wang et al. 2016b). Therefore, the direct combustion of carbon produced by char without prior gasification with Fe₂O₃ encouraged the application of chemical looping combustion of solid fuels (Rubel et al. 2009).

Although copper (Cu-) based oxygen carriers mostly demonstrate higher costs than Fe-based oxygen carriers and lower melting point (1084–1326 °C) than other oxygen carriers, the properties such as high oxygen carrier capacity, high durability, and low environmental impact of Cu-based oxygen carriers increase the interest for them in chemical looping technologies (Adánez et al. 2006; Yamaguchi et al. 2012). Based on the thermodynamic analysis, Cu-based oxygen carriers strongly support the feasibility of the chemical looping combustion process for solid fuels such as coal, biomass, and solid wastes (Cao et al. 2006). Studies into the reactivity of Cu-based oxygen carriers verified the combustion reaction of both gas and solid fuels with Cu-based oxygen carriers (Cho et al. 2004; Mattisson et al. 2009) because of their high reactivity, more extensive oxygen transfer, and releasing capacities (Cao and Pan 2006). For example, the primary Cu-based synthetic oxygen carrier, CuO, demonstrated high reduction and oxidation so it can be described as an excellent oxygen carrier for solid fuel chemical looping combustion applications (Siriwardane et al. 2009). It has also been demonstrated that CuO can be fully converted into Cu in a reduction process, either through the combustion of solid fuels or the indirect combustion of pyrolysis and gasification products of solid fuel (Cao et al. 2006; Güleç et al. 2023b, 2023c). The combustion and oxidation performance of Cu-based natural ores as an oxygen carriers

is not as good as synthetic CuO oxygen carriers due to the low active species concentration in ores (Tian et al. 2013). Despite the higher oxygen capacity of Fe-based natural ores, Cu-based natural ores showed faster reaction rates than Fe-based natural ores in the chemical looping combustion of methane (Tian et al. 2013). No carbon deposition (Adánez et al. 2006; Cao et al. 2006), little or no agglomeration (Cho et al. 2004; Adánez et al. 2006) and no surface sintering (Siriwardane et al. 2009) after combustion and oxidation of Cu-based oxygen carriers.

Nickel (Ni-) and manganese (Mn-) based oxygen carriers are also suitable for chemical looping applications since both demonstrate relatively high melting points, high oxygen capacity and low agglomeration (Leion et al. 2009; Chiu and Ku 2012). Though they have lower oxygen transfer capacities than Cu-based oxygen carriers (Cao et al. 2006), they show better reactivity (Cho et al. 2004; Chiu and Ku 2012). On the other hand, Mn₂O₃ and NiO showed a lower combustion rate for solid fuels compared to Cu-, Fe- and Co-based oxygen carriers. Carbon deposition, cost and toxicity of the carriers are still significant problems to be solved with Ni-based carriers (Mattisson et al. 2003; Cho et al. 2005; Chiu and Ku 2012). Carbon deposition of Ni-based oxygen carriers may be addressed using the correct Ni concentration on an oxygen carrier (Cho et al. 2005; Sedor et al. 2008; Rubel et al. 2009).

Cobalt (Co-) based oxygen carriers showed excellent performance in chemical looping biomass gasification thanks to their superior properties such as high oxygen capacity, mechanical strength, high melting point, and thermal stability (Cao et al. 2006). Oxygen release capacity and thermal stability make Co a preferred oxygen carrier compared to Mn and Cu (Aghabarnejad et al. 2014), while the high cost and toxicity reduce its application in chemical looping applications (Aghabarnejad et al. 2014). In addition to CoO oxygen carrier, both CuO- and NiO-based oxygen carriers were illustrated to be the optimal oxygen carriers for the development of the chemical looping combustion of solid fuels in terms of thermodynamic analysis and physical properties of the oxygen carriers (Cao et al. 2006).

Combined oxygen carriers such as CoFe₂O₄, CuFe₂O₄, and Fe-Cu/Mg showed better reaction activity (Wang et al. 2010, 2013, 2014). A new combined oxygen carrier, Fe45Cu15M40, which consists of 45% of Fe₂O₃, 15% of CuO and 40% of MgAl₂O₄ was developed by some researchers (Wang et al. 2010), which maintained a relatively high activity and stability for long reduction and oxidation cycles. This also showed high reactivity in a pressurised chemical looping combustion unit for 15 h and provided for the high conversion of the fuel. Furthermore, the catalyst showed resistance towards agglomeration and fragmentation. The desired reaction superiority of CoFe₂O₄ over single reference oxides Fe₂O₃ and CoO has been reported by some

researchers (Wang et al. 2014). The principal reduction of CoFe_2O_4 is to Fe_3O_4 and Co, which was also verified by thermodynamic simulations.

Gasification experiments with oxygen carriers generally performed better than inert sand (Di Giuliano et al. 2021). The type of oxygen carriers, composition, and reduction steps have a significant impact on syngas production and its composition. As the highest hydrogen yield (23.1 mmol/g biomass) was produced at the optimum ratio of Fe/Ca (1:1 of $\text{Ca}_2\text{Fe}_2\text{O}_5$) at 800 °C thanks to one step reduction and oxidation of $\text{Ca}_2\text{Fe}_2\text{O}_5$ (Hu et al. 2020). LD slag also known as steel converter slag consists of Ca, Mg, Al and Mn besides iron in a complex matrix of phases, which makes it a cheap and potential oxygen carrier for the chemical looping gasification processes. The presence of CaO makes LD slag a promising oxygen carrier candidate for biomass chemical looping gasification to achieve syngas with a high H_2/CO ratio (Hildor et al. 2020), where the ratio of H_2/CO was 28 at a low temperature (820 °C). This also could be attributed to the extra hydrogen production via catalysed WGS reactions and water splitting reactions. However, H_2 production decreased due to the low stability of the Fe–Ca oxygen carrier (Hu et al. 2020), as the bimetallic structure of oxygen carriers was decomposed by the accumulation of Si in the ash composition of biomass and formed CaSiO_3 and Fe_2O_3 following a few cycles (Hu et al. 2020).

Characterisation of biomass and oxygen carriers

Biomass/biochar, as a fuel source, and metal oxides, as oxygen carriers, possess unique physicochemical properties, which influence their behaviour and performance in chemical looping applications. Understanding the relationship between biomass/biochar and metal oxide properties and their response to chemical looping combustion and gasification is crucial for optimising the efficiency and effectiveness of these technologies and developing strategies to mitigate challenges associated with conversion technologies.

Proximate, ultimate, chemical analyses

Biomass/biochars tend to demonstrate different moisture, volatile matter, fixed carbon, and ash content due to the variety of nature of the geographical locations. Similarly, the elemental composition (carbon, hydrogen, oxygen, nitrogen, sulphur) also demonstrates significant differences. Identifying these differences provides insights into their thermal behaviour, combustion characteristics, reactivity and kinetics (Güleç et al. 2021b, 2022b, 2022d). Higher volatile matter content and reactivity of biomass can facilitate efficient gasification in chemical looping gasification. Since volatiles

can directly participate in a series of catalytic reactions, undergoing different types of transformations (Ji and Shen 2022). During the chemical looping gasification of algae, the volatiles reduce the Ni^{3+} to Ni^{2+} , which promotes a process of charge compensation, leading to the formation of oxygen vacancies (Fino et al. 2003). The introduction of Fe in oxygen carriers enhances the cleavage of pyrolytic volatiles and promoted the generation of CO and CH_4 (Zou et al. 2018). On the other side, biochar having low volatile content caused a decrease in the CH_4 concentration through biochar gasification stage (Abdalazeez et al. 2022). Additionally, biochars with higher fixed carbon content and lower volatile matter can have enhanced gasification performance in chemical looping gasification.

In addition to these properties, the chemical composition of biomass/biochar namely cellulose, hemicellulose, lignin, and inorganic components including alkali and alkaline earth metals, influences their combustion and gasification behaviours. Guo et al. (2023) investigated the chemical looping combustion behaviours and mechanisms of cellulose using Fe-based oxygen carriers (Guo et al. 2023). The chemical looping combustion of cellulose shows three reduction stages; (1) depolymerisation of cellulose into active cellulose, volatile analysis out and combustion, (2) rearrangement carbonisation of heavy components, and (3) fixed carbon combustion (Guo et al. 2023). Lignin shows lower gas production and higher tar production compared to cellulose and hemicellulose under the same gasification conditions (Yu et al. 2018). Higher lignin content in biomass can hinder reactivity, while higher cellulose content and lower lignin content may exhibit improved combustion reactivity during chemical looping combustion. However, the syngas ($\text{CO} + \text{H}_2$) generations would vary based on the applied metal oxides, therefore, lignin can also provide a decent amount of gasification with metal oxides. In a comparison study of bimetallic metal oxide application to lignin gasification, syngas production is favour with the following order $\text{BaFe}_2\text{O}_4 > \text{CaFe}_2\text{O}_4 > \text{BaMn}_2\text{O}_4 = \text{CaMn}_2\text{O}_4 >$ lignin only (Wu et al. 2020). The highest capacity of syngas was determined as 0.63 m^3 per kg lignin waste (Fig. 5). As in conventional combustion, estimation of the unburnt carbon (Golgiyaz et al. 2023b) is also significantly important in the chemical looping combustion of biomass and chemical looping gasification of biomass process optimisation.

Ash and alkali-alkaline earth metals

The presence of alkali and alkaline earth metals in biomass/biochar can influence the reactivity of oxygen carriers in chemical looping combustion and gasification and affect ash-related issues, such as agglomeration, fouling, and slagging, as demonstrated in Fig. 6 (Di Giuliano et al. 2020). A summary of complex reaction pathways for the

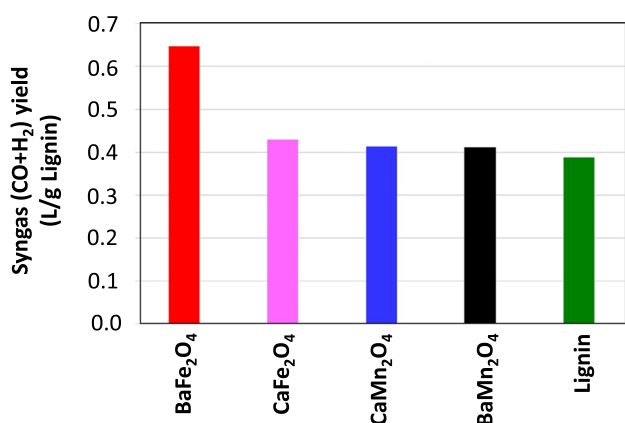


Fig. 5 The syngas product yield for the gasification of lignin with and without bimetallic oxygen carriers, reprinted with permission of Elsevier from (Wu et al. 2020)

thermal conversion of automotive shredder residue in a chemical looping technology is provided in Fig. 6 based on experimental results and thermodynamic equilibrium prediction (Stanicic et al. 2021). The reaction mechanisms

include drying, devolatilisation, char burnout, the interaction between ash and metal oxides, oxidation and reduction mechanisms of the metal oxides, and formation of metal chlorides, in addition to the total amount of metals released depending on several factors such as temperature, reduction potential but also the Cl- and S-chemistry (Stanicic et al. 2021).

Furthermore, at reduced levels of biomass carbon in the system, the combination of Fe- based oxygen carrier and ashes led to the creation of compounds including MgFe₂O₄, Fe₂SiO₄, and CaFe₂O₄ (in Fig. 7) (Cheng et al. 2020). It was observed that these compounds posed greater difficulty for reduction compared to Fe₂O₃. This kind of interaction between metal oxides and ash significantly decreases the activity of metal oxides. The accumulation of Si from biomass ash over Ca₂Fe₂O₅ decreases the hydrogen yield in multicycle chemical looping gasification due to the decomposition of Fe–Ca bimetallic structure and formation of CaSiO₃ and Fe₂O₃ after the 3 cycle (Hu et al. 2020). Si, Mg and Ca migrate from the used bed material to the char during the process and the alkali released from the used bed materials is absorbed by the char when they are mixed

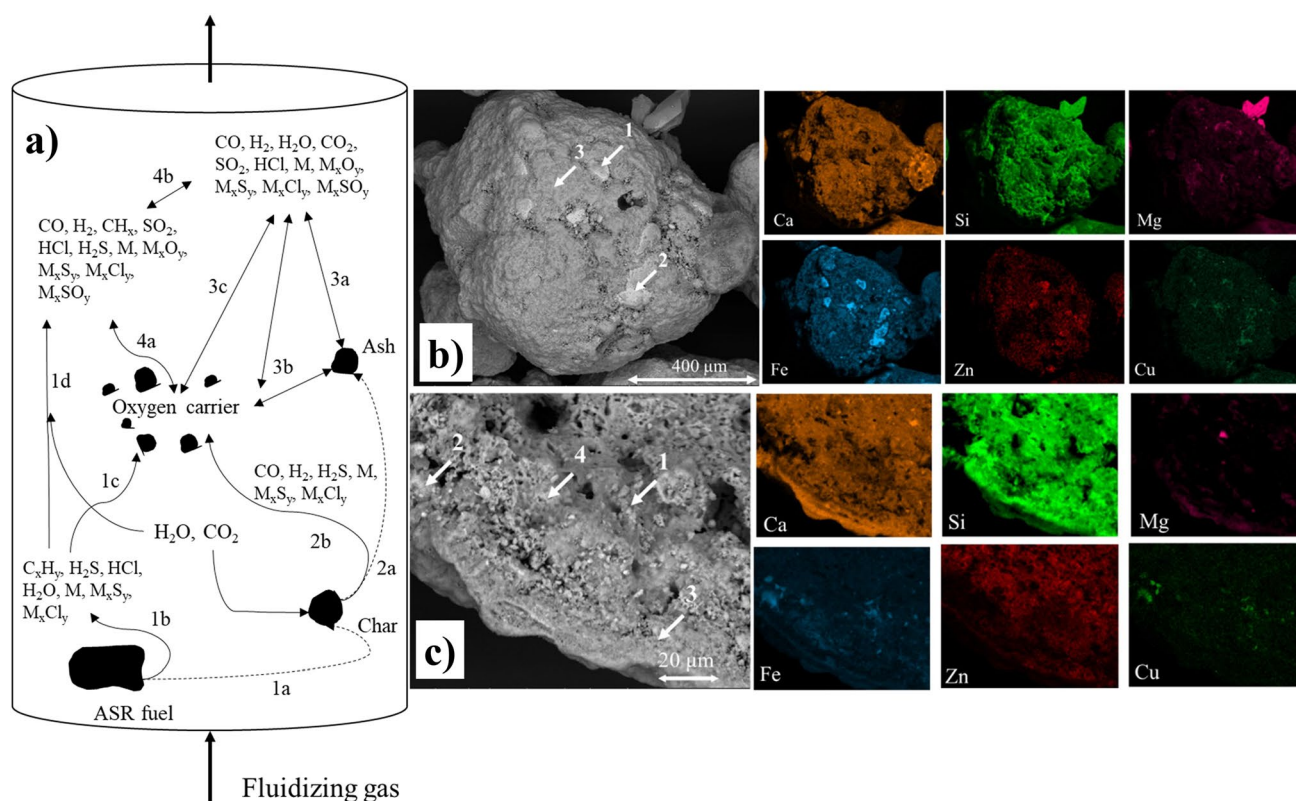


Fig. 6 **a** Possible pathways for the elements Cu, Zn, Sb and Pb (also represents as M) compiled based on experimental results and thermodynamic equilibrium prediction as performed via automotive shredder residue (ASR), further mechanistic explanation can be found in (Stanicic et al. 2021). **b** Elemental mapping of the surface of a parti-

cle in the bottom ash (BA) from day 13 and **c** mapping of a particle, exposing its porous interior, obtained from different parts of the process on day 13, reprinted with permission of Elsevier from (Stanicic et al. 2021)

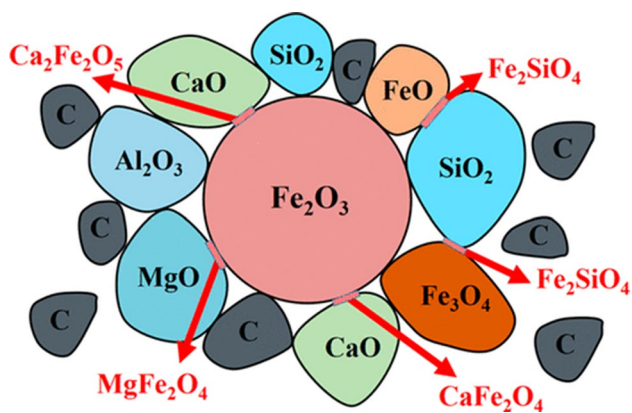


Fig. 7 Solid phase reaction between Fe-based oxygen carrier and ash, reprinted with permission of American Chemical Society from (Cheng et al. 2020). Carbon (C) represents the remaining part of solid fuel, iron oxide (Fe_2O_3) represents the oxygen carrier, iron(II, III) oxide (Fe_3O_4) represents the reduced oxygen carrier, silicon dioxide (SiO_2), aluminium oxide (Al_2O_3), magnesium oxide (MgO), and calcium oxide (CaO) represent the ash content. Bimetallic oxides (MgFe_3O_4 , CaFe_3O_4 , $\text{Ca}_2\text{Fe}_3\text{O}_5$, Fe_2SiO_4) represent the interaction of metal oxide with ash

as observed by scanning electron microscopy and energy dispersive X-ray spectroscopy analysis (Ge et al. 2022). In order to minimise the impact of biomass ash on the process, pretreatment was applied to biomasses such as torrefaction, leaching, and combination, which significantly improve the ash related characteristics (Lebedig and Müller 2022). Chlorine content was decreased by the application of torrefaction, but not the content of potassium. By changing the molar ratio of $\text{Si}/(\text{Ca} + \text{Mg})$ may affect the ash fusion; the higher the ratio, the higher the ash fusion temperature (Lebedig and Müller 2022).

Particle and surface analysis

Particle size distribution and surface area of biomass/biochar and metal oxides significantly impact the reaction kinetics and mass transfer processes during chemical looping combustion and chemical looping gasification. Fine biomass/biochar particles and higher surface area promote faster combustion/gasification reactions and enhanced reactivity. The chemical looping process consists of a high speed riser reactor, fluidised bed reactor, gas solid cyclone separator, low velocity bubbling bed, and loop seals inevitably led to the attrition of the oxygen carrier (Liu et al. 2022). The long-term applications of the chemical looping process and its conditions negatively impact the performance of oxygen carriers including abrasion, fracture, disintegration, and splitting (Liu et al. 2022). An average attrition rate of 0.16 wt%/h for the ilmenite in the application of chemical looping gasification of biomass was obtained, which corresponds to a lifetime of 630 h (Condori et al. 2021). While the

overall condition of the ilmenite particles was satisfactory, certain fragmentation became apparent on the outer surface of the particles. Consequently, during the operation, some particles experienced breakage, causing certain sections of their external shell to detach (Condori et al. 2021). The attrition behaviour of hematite particles was identified by the Gwyn kinetic equation fittings, which are jointly controlled by abrasion and fragmentation (Li et al. 2022). At higher temperatures, particles showed increased susceptibility to surface abrasion, whereas, during the reduction stage, fragmentation was more likely to occur compared to the oxidation stage (Li et al. 2022). The oxygen carriers show a high attrition rate in the early stage of the operation as a result of the detachment of fines, while the attrition rate remains stable at the later stage of the operation (Samprón et al. 2023).

Challenges

Chemical looping technologies are among the most promising BECCS options since they are considered to bring about the lowest energy and cost penalty for CO_2 capture. However, biomass feedstocks produce ashes with a high inorganic matter content, which can cause serious deactivation of the oxygen carriers used in the application of chemical looping combustion of biomass and chemical looping gasification of biomass processes (Di Giuliano et al. 2020). The application of chemical looping technologies to combustion and gasification can solve some of the major challenges faced in conventional biomass combustion and gasification. For example, biomass combustion has lower energy efficiency due to the aggressive alkali ash components although the ash content is relatively low (Zajac et al. 2018). However, chemical looping application could reduce this problem as oxygen carriers can absorb alkali metals, i.e. ilmenite can absorb potassium (Corcoran et al. 2018). Additionally, in the conventional gasification/combustion of biomass, volatile ash components, such as potassium and chlorine, can also be released during pyrolysis, which may leave the system in the gas flow. Low surface temperatures of heat exchangers downstream of the fuel reactor could be necessary to accommodate this. However, in chemical looping technologies, most of the process heat is extracted inside the air reactor instead of the fuel reactor. Therefore, the steam pipes are not supposed to be exposed to the biomass ash components and are not expected to be fouled easily (Lyngfelt 2020). Chemical looping combustion of biomass and chemical looping gasification of biomass is potentially a way to maintain high steam temperatures as well as reduce operational difficulties and costs by removing volatile ash components. As such, chemical looping combustion could also open up a broader

fuel diet for power production with high alkali fuels such as straw (Lyngfelt 2020).

The application of chemical looping combustion and gasification of biomass is relatively new and provides specific technical challenges that need to be addressed. The initial steps towards validating this technology require detailed experimental analysis on (1) Understanding the suitable type of biomass and biochar, i.e. What types of biomasses and biochar are mostly applicable for these technologies? (2) Suitable type of oxygen carriers, i.e. What type of oxygen carriers have the highest potential in chemical looping combustion and gasification of biomass? As some oxygen carriers demonstrate chemical looping oxygen uncoupling properties to release their oxygen due to thermal decomposition, some others do not release the oxygen themselves. (3) Understanding the chemical looping combustion and gasification of biomass process conditions, i.e. What are the optimised conditions of temperature, reactor type and configuration, fluidisation gas (CO_2 and H_2O), reaction mechanisms for the application of chemical looping combustion and gasification of biomass applications. (4) Interaction of ash oxygen carrier: How does the ash composition of particular biomass/biochar feedstocks and ratio impact the process and oxygen carrier? Although some oxygen carriers can absorb alkali metals in the ash, i.e. ilmenite can absorb potassium, the impact of these impurities on the oxygen carriers, their activity, and reusability has to be investigated carefully. (5) Ash and oxygen carrier separation: Potential separation technologies and applicability on solid ash oxygen carrier separation have to be developed for the application of chemical looping combustion and gasification of solid fuels. (6) Minimise the cost uncertainty of chemical looping combustion and gasification of biomass: a detailed cost analysis has to be processed in order to understand the real potential of these technologies, more experimental research on these technologies results in a detailed techno-economic analysis with a higher confident level.

Conclusion

Chemical looping combustion and gasification of biomass are two promising biomass-based capture technologies that have been implemented for the decarbonisation of several industries. Compared to other CO_2 capture technologies, both processes yield lower energy penalties and capture costs. However, the use of biomass feedstock during chemical looping processes leads to ash and char deposition that could ultimately deactivate the oxygen carrier. The present study presents state-of-the-art chemical looping combustion and gasification of biomass technologies together with the process challenges and future research directions. An overview of different oxygen carriers is discussed as well as their

advantages and limitations. Based on the literature overview, iron-based oxygen carriers are promising due to their low cost and environmental impact, improved mechanical strength and higher melting point compared. Copper based oxygen carriers are more expensive than Fe-based oxygen carriers and exhibit lower melting points (1084–1326 °C) than other oxygen carriers. In contrast, combined oxygen carriers such as CoFe_2O_4 , CuFe_2O_4 , and Fe–Cu/Mg showed improved reaction activity. Chemical looping technologies are relatively new technologies and face challenges related to the screening and selection of a suitable type of biomass and biochar as well as the underlying reaction mechanism. It should be mentioned that while some oxygen carriers showed chemical looping oxygen uncoupling properties, some others do not release the oxygen themselves. Therefore, it is very important to understand the impact of process conditions, the interaction of ash oxygen carriers and reaction mechanism during chemical looping technologies.

Author contribution FG contributed to the data collection, project administration, funding, formal analysis, visualisation, investigation, writing—original draft, writing—review and editing; JAO was involved in the formal analysis, validation, writing—original draft, writing—review and editing.

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Availability of data and materials All relevant data are within the manuscript and available from the corresponding author upon request.

Declarations

Conflict of interest The authors have not disclosed any competing interests.

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