

Chemical looping combustion: Advantages, disadvantages, opportunities and challenges

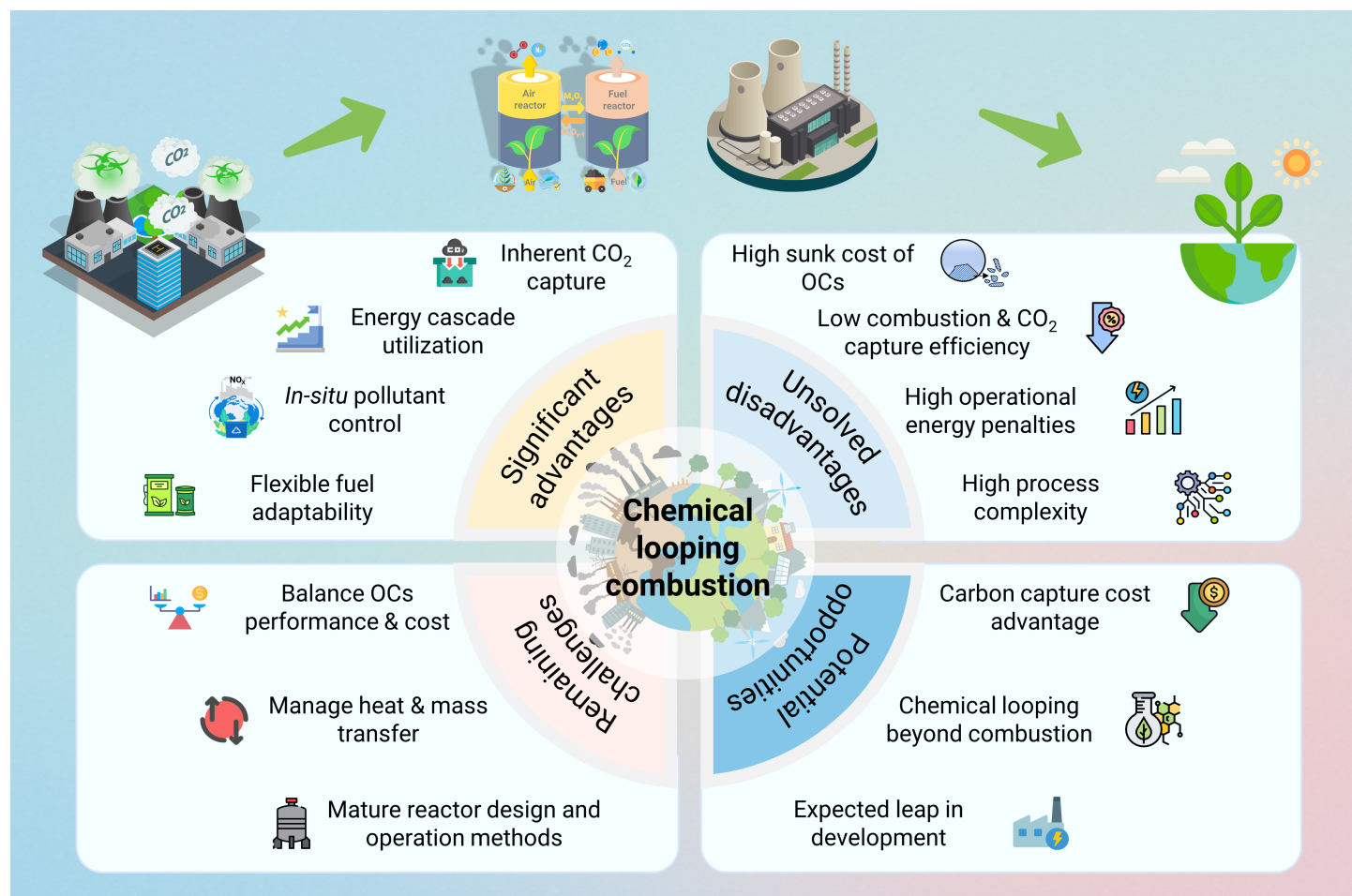
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GRAPHICAL ABSTRACT



PUBLIC SUMMARY

- Chemical looping combustion (CLC) offers inherent CO₂ capture and cascade energy utilization.
- Key challenges include cost-performance balance of oxygen carriers and reactor optimization.
- Commercialization is hindered by operational complexities and efficiency gaps.
- Advancing CLC requires cost-effective oxygen carriers and standardized reactor designs.

Chemical looping combustion: Advantages, disadvantages, opportunities and challenges

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Chemical looping combustion (CLC) has garnered significant attention for its inherent CO₂ separation and efficient energy utilization, making it a leading low-energy CO₂ capture technology. Despite substantial progress, from studies on oxygen carriers (OCs) to pilot-scale applications, further research is essential for commercial deployment. This review examines the current status of CLC, highlighting its advantages, disadvantages, unresolved challenges, and opportunities. Key challenges include balancing OC cost and performance, optimizing heat and mass transfer, accelerating rate-limiting reactions, and improving reactor design and operation. The analysis reveals that while CLC demonstrates promising efficiency and scalability, its full commercialization is hindered by operational complexities and efficiency gaps. To advance CLC, this study recommends prioritizing the development of cost-effective, high-performance OCs and standardized reactor design methodologies through interdisciplinary collaboration and advanced computational tools.

INTRODUCTION

It is widely recognized that carbon dioxide emissions from the combustion of fossil fuels are the primary driver of global warming. The average peak of CO₂ concentration in the atmosphere has reached 421.37 ppm, the highest recorded since the Industrial Revolution. Over the long term, fossil fuels are expected to continue being a predominant energy source globally.¹ To address this issue, it is vital to achieve low-carbon use of fossil fuels or even negative carbon use of carbon-neutral fuels.

Carbon capture, utilization, and storage (CCUS) technology is thought to be a crucial choice that can lead to a notable decrease in conventional fossil energy industries. According to the model predictions of the Intergovernmental Panel on Climate Change (IPCC), CCUS technology is considered indispensable for achieving the 1.5 °C climate goal, and the abatement cost would increase by 138% without CCUS.² China has the highest carbon emissions globally, with its energy consumption predominantly reliant on coal.³ CCUS provides a feasible option that allows for the control of CO₂ emissions as well as bolstering the coal-centric energy framework of China. Nevertheless, the implementation of CCUS has been hindered by the high expense of current carbon capture techniques.

Chemical looping combustion (CLC), a novel way for the low-carbon utilization of fossil fuels, has the advantage of inherent CO₂ separation, which eliminates the need for energy-intensive separation steps required in conventional CO₂ capture technology.⁴ This positions CLC as a potential technology for scalable, low-cost CCUS systems. After decades of development, the CLC process has been demonstrated in units of different sizes, from bench scale to MW-scale pilot plants.^{5,6} While substantial advancements have been achieved in CLC development, the commercialization of CLC still faces bottlenecks. Previous review articles have focused on research progress in oxygen carrier (OC) materials^{7,8} or reactor configurations,^{9,10} providing valuable insights into material selection and equipment design for researchers. Readers can become familiar with these concepts through some recent reviews.^{11–14} Nevertheless, these reviews have seldom systematically explored the key challenges inherent in CLC technology and potential solutions.

Notably, the purpose of this review is not to reiterate existing literature related to CLC technology but rather a summary of relevant perspectives from a broader, overarching viewpoint. As shown in Figure 1, this paper delves into the potential opportunities and existing challenges of CLC technology, starting from an analysis of its advantages and disadvantages. It

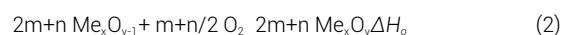
offers a dialectic analytical framework, proposing potential solutions to provide valuable insights for the commercialization of CLC technology and further increase the feasibility of large-scale CCUS technology implementation.

CHEMICAL LOOPING COMBUSTION

Basic principle

As illustrated in Figure 2A, the CLC system comprises two interconnected reactors, the air reactor (AR) and the fuel reactor (FR), which are atmospherically isolated. Instead of gaseous oxygen, it uses an OC to transfer lattice oxygen between the two reactors for fuel conversion.¹⁵ In the FR, the active component of the oxidized OC reacts with the fuel, transferring lattice oxygen to the fuel for combustion (Equation 1). In the absence of air dilution, the effluent from the complete combustion of FR consists exclusively of CO₂ and H₂O. As a result, no additional separation equipment is required. High-purity CO₂ stream can be obtained through steam condensation, and the resulting CO₂ can be further utilized for other applications.^{16–18} Subsequently, the reduced OCs are sent to the AR, where they are regenerated to its oxidized state by air oxidation and releases heat (Equation 2). The OC is then sent back to the FR for the next redox cycle.

In addition, based on thermodynamic analysis, the total reaction heats of the reduction (Equation 1) and oxidation (Equation 2) in the CLC process is equal to the reaction heat of the conventional combustion reaction (Equation 3).¹⁹ However, CLC decouples the co-feeding process of oxygen and fuel into two independent processes, achieving cascaded energy utilization without increasing the enthalpy of the combustion reaction, and the exhaust gas after combustion can be coupled with other thermal systems to enhance overall energy efficiency.^{20,21}



Development status

The historical development roadmap of research related to chemical looping is presented in Figure 2B. The origins of chemical looping can be attributed to the “steam-iron” technique for H₂ production proposed by Lane et al.²² in the early 20th century and the patent that produces high-purity CO₂ through the redox reaction involving metal oxide in 1954.²³ In 1983, Richter and Knoche²⁴ suggested substituting the conventional one-step air combustion process with two distinct redox sub-steps in order to enhance the exergy efficiency in power plants, which also served as the prototype for modern CLC. It wasn't until 1987 that Ishida et al.²⁵ formally presented the concept of CLC. Furthermore, Ishida and Jin²⁶ explored that the CLC process had the advantage of inherent CO₂ separation and could reduce NO_x emissions. Given the increasing global concern over climate change issues, CLC technology has entered a rapid development stage. In this process, recognizing the critical role of reactor selection in successful CLC implementation, researchers have proposed various reactor designs and conducted extensive experiments and trials.

Three kinds of reactor concepts have been proposed for the CLC process: the dual fluidized bed reactor, packed bed reactor, and the rotating reactor, which correspond to the loop of OCs, the reactor, and the reaction atmo-

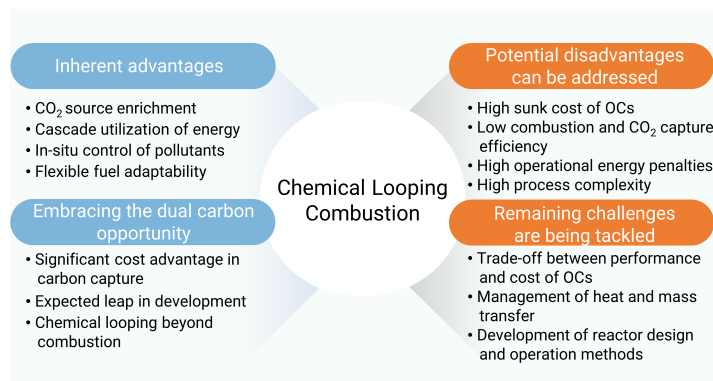


Figure 1. The graphical summary of this review's framework.

sphere during the reaction process, respectively (Figure 2C). Early in this century, Lyngfelt et al.^{27,28} from Chalmers University of Technology (CUT) in Sweden designed, constructed, and successfully operated a 10 kW_{th} CLC reactor using a dual fluidized bed, proving the feasibility of CLC from paper concept to practical application. The dual fluidized bed shares certain similarities with traditional circulating fluidized bed power generation systems, making it suitable for large-scale, high-capacity industrial applications. During this period, Noorman et al.²⁹ proposed using a fixed-bed reactor for gas-fueled CLC, which is compact and well-suited for high-pressure operations. It offers an alternative approach to the practical engineering implementation of CLC technology. Moreover, novel reactor designs, such as rotating reactors,³⁰ have also been explored for CLC applications, though their feasibility remains constrained by various limitations. The variety of reactor designs enhances the fuel adaptability of CLC, making it comparable to traditional combustion technologies. While early CLC research primarily focused on gas fuels, recent years have seen increasing attention on coal and biomass.

As depicted in Figure 2D, there are typically three ways to achieve CLC of solid fuel: Syngas-CLC, *in-situ* Gasification CLC (*IG*-CLC), and Chemical Looping with Oxygen Uncoupling (CLOU).³¹ The process of Syngas-CLC involves the pre-gasification of solid fuel followed by utilizing the generating syngas as fuel to react with OC. In the *IG*-CLC process, the solid fuel is introduced directly into the FR and undergoes *in-situ* gasification in the presence of a gasifying agent (H₂O and/or CO₂), followed by the combustion of the gasification products.^{32,33} However, the rate of char gasification is up to 100 times more slowly than combustion, resulting in slow overall reaction rates. To address these issues, Mattisson et al.³⁴ proposed the concept of CLOU, in which OC (such as Mn₂O₃ or CuO) releases gaseous oxygen at high temperatures, which subsequently reacts with a fuel. The proposed approach achieves better combustion and carbon capture efficiency by eliminating sluggish gasification.

The features of OC are essential in both the design and performance of the CLC system. In the last few years, researchers have dedicated substantial endeavours to advancing the development of OC.³⁵ The key criteria for ideal OCs include redox reactivity, oxygen transport capacity, mechanical properties, cost-effectiveness and availability, and contaminant resistance.³⁶ OCs can be mainly divided into synthetic materials and natural ores/industrial wastes. Synthetic OCs include single-component or mixed oxide OCs such as Ni-based, Cu-based, Fe-based, Mn-based,^{37–39} sulfate-based,⁴⁰ and perovskite-based OCs.⁴¹ Natural OCs are divided into natural ores,⁴² modified ores,⁴³ and industrial waste OCs.⁴⁴ Although metal oxides have good reaction activity, their cyclic stability is poor, necessitating the use of inert carriers (e.g., Al₂O₃, SiO₂, TiO₂, and ZrO₂) to improve their thermal stability and mechanical strength.^{45–47} Obviously, the preparation method of OCs significantly affects the dispersion of the active phase on the carrier and the mixing degree at different scales. The primary methods for preparation include mechanical mixing, wet impregnation, spray drying, sol-gel, freeze granulation, and hydrothermal synthesis.^{48–50} Although many efforts have been made to study the performance of OCs, these studies are mainly concentrated on a batch fluidized bed reactor or even the static thermogravimetric analyzer (TGA)/fix-bed conditions, which are not adequate to reflect abrasion and

fragmentation, sintering and agglomeration, and poisoning and deactivation of OCs that may occur in an actual CLC process. Therefore, it is crucial to do experiments in actual CLC circumstances, such as within a continuously operated dual fluidized bed reactor. Currently, most OCs used in existing CLC pilot plants are natural ore OCs,^{51–53} whereas synthetic OCs have not been sufficiently tested in continuous units due to high cost.^{54,55}

INHERENT ADVANTAGES

CO₂ source enrichment

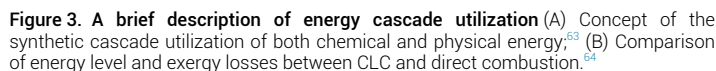
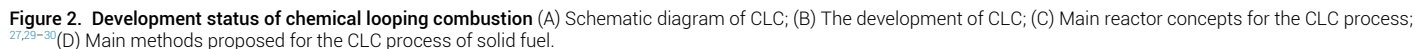
CCUS allows for the continued use of fossil fuels while substantially mitigating CO₂ emissions. However, CCUS demonstration projects in China still suffer high energy consumption and costs. The most expensive component of CCUS is the CO₂ capture process, which accounts for 60 to 70% of overall expenditures and up to 90% when considering compression.⁵⁶ The expense of CO₂ capture is mainly determined by the emission source and the purity of CO₂, where higher concentrations of CO₂ result in lower cost and energy penalty for capture. Nonetheless, it should be noted that the proportion of high-concentration point source emissions in China is relatively minimal. The CLC process allows for fuel oxidation without the need to blend air with combustion products, resulting in flue gases devoid of N₂, primarily composed of CO₂ and water vapour. Following steam condensation, nearly pure CO₂ is acquired.

Because of its ability to *in-situ* capture high-concentration CO₂ during combustion, additional gas separation equipment can be eliminated, reducing the capture cost. Lyngfelt et al.⁵⁷ studied the expenses associated with a 1000 MW_{th} boiler for CLC in solid fuels. The overall expense associated with CO₂ capture was determined to be 22 \$/tonne CO₂ avoided, falling between 18–29 \$/tonne, significantly less than the expenditures for pre-combustion capture (\$28–41/tonne CO₂), oxy-fuel combustion (\$36–67/tonne CO₂), and post-combustion capture (\$36–53/tonne CO₂).⁵⁸ A techno-economic assessment of typical 2×300 MW CFB and CLC-based coal-fired units in China was carried out by Zou et al.⁵⁹ According to the analysis, the carbon reduction cost of the CLC power plant is ¥72.15/tonne CO₂, which was lower than that of IGCC technology, oxy-fuel combustion technology, and CFB technology (¥85.55–342.19/tonne CO₂).⁶⁰ Those results demonstrate that the CLC technology seems more efficient, cost-effective, and environmentally benign due to CO₂ source enrichment.

Energy cascade utilization

For a long time, the efficient use of energy has been centred on the transition of physical energy. In fact, the most significant degradation of exergy in thermal cycles takes place during the combustion process.⁶¹ When fuel is burned directly, its valuable chemical energy is transformed into thermal energy, which reduces the overall efficiency of converting fuel into electricity in power systems. This procedure leads to a larger irreversible loss, which can account for 50% of the overall losses in the system.⁶² The circulation of OCs is crucial in facilitating energy release during the CLC process. Unlike traditional methods that directly convert chemical energy into physical energy, this approach employs a sequential energy release mechanism. In this process, the chemical energy of the fuel is first transferred to the OCs and then converted into thermal energy.

This innovative technique, as opposed to direct combustion, enables a more efficient utilization of chemical energy before the combustion stage, minimizing the irreversible energy loss during the conversion to heat. As shown in Figure 3A, a notable difference ($A_{ch1}-A_{Th}$) exists between chemical and physical energy levels during direct combustion.⁶³ Integration of the CLC method with the chemical reaction procedure enables a gradual use of chemical energy derived from fuel, facilitating the transition from energy level A_{ch1} to A_{ch2} . This approach reduces the energy level disparity between chemical and physical energy during CLC combustion to ($A_{ch2}-A_{Th}$), thereby minimizing exergy losses. Figure 3B compares energy level and exergy losses in CLC and direct combustion for coal combustion processes.⁶⁴ The chemical exergy levels of CLC and direct combustion are 0.85 and 1, respectively, demonstrating that the chemical exergy level in CLC is lower and closer to that of the heat source. This allows a greater proportion of chemical exergy to be converted into physical energy. Thus, exergy losses in CLC are reduced by approximately 10.35% compared to direct combustion. Additionally, CLC



Dioxins emission. Previous research has summarized that PCDD/Fs may be generated through two main mechanisms:^{65–66} (1) precursor conversion, where precursors undergo reactions such as chlorination, condensation, and

The de novo synthesis and precursor conversion of PCDD/Fs can be controlled owing to the O_2 -free atmosphere and *in-situ* dechlorination characteristics of the CLC technology. Bi et al.⁷⁴ discovered that the oxidation and cracking of macromolecular carbon and the Deacon reaction were inhibited during the CLC process due to no contact between fuel and O_2 . Through thermodynamic simulation, Hua et al.⁷⁵ found that the activity of lattice oxygen in the Deacon reaction was reduced compared to gaseous oxygen, which suppressed the generation of PCDD/Fs by reducing the chlorine source from the source. Zhao et al.⁷⁶ first applied CLC technology to control the generation of PCDD/Fs while incinerating plastic waste. The analysis of 17 toxic PCDD/Fs (Figure 4A) in the exhaust gas showed that, compared to air incineration, the *in-situ* dechlorination using CaO-modified Fe_2O_3/Al_2O_3 as OC (with an 82% reduction in Cl_2 yield) led to a remarkable decrease of 75% and 88% in toxicity equivalent and generation of PCDD/Fs, respectively. The suppression mechanism of PCDD/Fs by CLC can be manifested as follows:⁷⁷ The absence of oxygen in the combustion process hinders the creation of PCDD/Fs through heterogeneous de novo synthesis at low temperatures, while the synergistic effect of an O_2 -free atmosphere and CaO dechlorination ($CaO + HCl \rightarrow CaCl_2$) controls the generation of Cl_2 , thereby indirectly weakening the chlorination in the high-temperature gas-phase synthesis pathway.

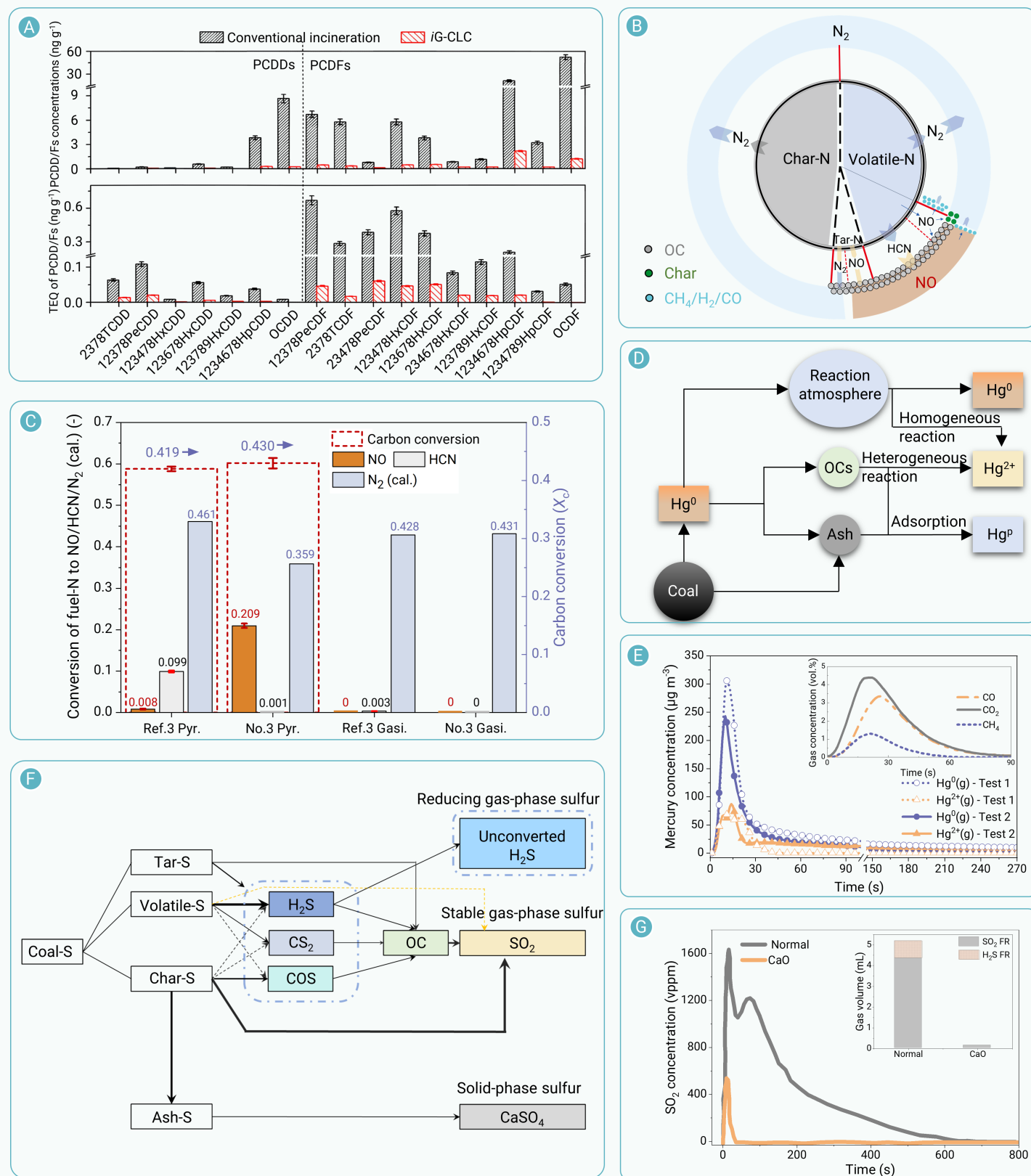


Figure 4. The pollutants *in-situ* control performance of CLC (A) The concentration distribution of 17 toxic PCDD/Fs congeners;⁷⁷ (B) Fate of nitrogen in volatiles, char, and tar of coal during iG-CLC of coal;⁷⁹ (C) Conversion of fuel-N to NO, HCN, and N₂ with (No.3) and without (Ref.3) OCs;⁷⁹ (D) Mercury migration and transformation process during a coal CLC;³⁵ (E) Hg⁰(g) and Hg²⁺(g) concentrations during the coal pyrolysis process for the blank experiment (test 1) and the CLC experiment (test 2);³⁷ (F) Sulfur migration and transformation process during a coal CLC;⁹¹ (G) SO₂ evolution of high-sulfur coal during the CLOU process with and without CaO addition.⁹⁴

and impeding precursor conversion.

Nitrogen emission. The primary sources of nitrogen oxides (NO_x) in coal combustion processes are as follows: (1) thermal NO_x, which is produced

when N₂ in the air is oxidized at temperatures above >1300 °C; (2) prompt NO_x, which arises from the reaction between N₂ in the air and hydrocarbon groups present in the fuel; and (3) fuel NO_x, which refers to the generation of

NO_x resulting from the oxidation of N-containing compounds within the fuel. Since the CLC process is conducted at a relatively mild temperature, there is almost no generation of thermal NO_x .⁷⁸ Generally, HCN, NH_3 , and HNCO serve as primary precursors of NO_x during the pyrolysis and gasification stages. The absence of air in the FR allows them to be converted by gas-solid heterogeneous reactions with OC particles. Compared to the conventional combustion process, this results in a markedly diminished oxidation of the reaction atmosphere.

For iG-CLC , fuel NO_x is solely responsible for the formation of NO_x . Ma et al.⁷⁹ investigated the fate of fuel-nitrogen during iG-CLC of coal using a two-stage fluidized bed reactor. It found that nitrogen compounds, primarily HCN, are released during the coal pyrolysis stage, while no NO_x is detected during the subsequent char gasification stage (Figure 4B). A Fe-based OC effectively converts HCN to NO or N_2 , with a CO_2 -rich atmosphere suppressing NO formation (Figure 4C). Song et al.^{80–81} performed a coal CLC process using Ni-based OC in a 1 kW_{th} CLC unit. The results showed that N_2 is the sole product in the FR, and no NO and NO_2 were detected. This phenomenon can be attributed to the reduction of the atmosphere generated during fuel devolatilization, which was conducive to reducing NO_x to N_2 . As for the AR, only the formation of NO was found. During the CLOU process, Vega et al.⁸² discovered that the N element present in the coal was still released mainly as N_2 in the FR, with approximately 20% being converted to NO. However, it was asserted that the NO concentration detected for both combustion modes stayed below the current legal limit for NO_x emissions in power plants.⁸³ In this case, CLC can effectively convert fuel NO_x to N_2 while also inhibiting the *in-situ* formation of NO_x , which allows compliance with the national NO_x emission standard without necessitating denitrification apparatus at the FR outlet.⁸⁴

Mercury emission. Mercury (Hg), a prominent pollutant emitted during coal combustion, poses significant risks to human health and the ecological environment. CLC will unavoidably result in Hg contamination due to the existence of Hg in coal.⁸⁵ As shown in Figure 4D, when coal is pyrolyzed and ignited during the CLC process of coal, the majority of Hg released is in the form of Hg^0 . Following the cooling of the flue gas, some of the Hg^0 reacts with the components of the flue gas to generate oxidized mercury (Hg^{2+}), and part of the Hg^0 is adsorbed onto fly ash or OCs surface as the state of particulate-bound mercury (Hg^{p}).⁸⁶ Therefore, the forms of Hg involved in the CLC process of coal mainly encompass Hg^0 , Hg^{2+} , and Hg^{p} . Hg^0 is highly volatile and poorly soluble in water, making it difficult to be effectively removed by existing pollution control devices. Therefore, effectively controlling Hg^0 in flue gas is key to addressing the issue of mercury pollution.

Ma et al.⁸⁷ examined the transformation of mercury during the process of coal pyrolysis and char gasification in the iG-CLC of coal. As can be seen in Figure 4E, although OC had a negligible impact on the release rate and quantity of mercury, it promoted the oxidation of Hg^0 to Hg^{2+} . This phenomenon can be attributed to two factors: (1) the sufficient lattice oxygen in the OC providing active sites for the oxidation of Hg^0 and (2) the oxidation of reducing volatile fractions and gasification products by the OC attenuating the inhibitory effect of reducing gases on the oxidation of Hg^0 . The Hg^0 removal efficiencies vary due to differences in the oxygen release capabilities of various OCs and their mercury adsorption properties in the reduced state. Perez et al.⁸² investigated mercury emissions during coal combustion using a novel Cu-Fe OC in the CLOU system. The results demonstrated that enhanced Hg oxidation in CLOU promoted its retention as Hg^{p} in ashes, resulting in a lower amount of gaseous Hg in CLOU (18.6%) than iG-CLC (31.5%) in the FR exit flow. Ni et al.⁸⁸ investigate the efficiency of Hg^0 oxidation using various OCs in the CLC of coal. It was obtained that the ranking of OCs based on Hg^0 oxidation efficiency is: $\text{CaSO}_4 > \text{Co}_3\text{O}_4 (\text{Mn}_2\text{O}_3) > \text{Fe}_2\text{O}_3 > \text{CuO} (\text{CeO}_2) > \text{SiO}_2 (\text{Al}_2\text{O}_3)$.

Sulfur emission. The emission of sulfur released from coal during CLC primarily occurs in the gas phase, such as H_2S , COS, CS_2 , and SO_2 , while the rest is fixed in coal ash as sulfates (e.g., CaSO_4) or reacts with OC to form metallic sulfides (e.g., Ni_3S_2 , Cu_2S , and FeS). Therefore, the performance of OCs is greatly impacted by the amount of sulfur in coal, which reduces CO_2 purity and carbon capture efficiency at the outlet of the FR.⁸⁹

One approach to remove sulfur-containing pollutants involves converting them into SO_2 within the FR, followed by flue gas treatment. Mendiara et al.⁹⁰ discovered that during iG-CLC of Spanish lignite, OC can partially oxidize the

reducing sulfur-containing gases, generating predominantly H_2S and SO_2 within the FR. Subsequently, upon char combustion in the AR, unconverted sulfur is primarily released as SO_2 . In addition, approximately 75% of sulfur present in coal is released in the gaseous phase, while the remaining 25% becomes fixed within the ashes. Ma et al.⁹¹ investigated the transformation of sulfur in pyrolysis products and char in iG-CLC . As shown in Figure 4F, it was found that 1/4 of the sulfur was found to be preserved in coal ash as a CaSO_4 component, while the remaining sulfur was mainly in the gaseous phase of SO_2 and trace amounts of unconverted H_2S , with almost no release of COS and CS_2 .

An alternative method involves transforming S into sulfide/sulfate, followed by its solidification in OC or ash. Teyssié et al.⁹² investigated the influence of limestone addition in an ilmenite bed for CLC. They found that the average concentration of SO_2 in the flue gas outlet dropped by 26% after adding CaO. Cuadrat et al.⁹³ studied the impact of adding limestone to ilmenite as an OC in a 10 kW_{th} CLC pilot, and no detectable levels of SO_2 were observed in the FR after limestone addition. Tian et al.⁹⁴ discussed the sulfur evolution behaviour of high-sulfur coal using copper ore as an OC during the CLOU process. It was found that sulfurous gases were mainly generated during the process of OC reduction, while a minor fraction of SO_2 was produced during the OC oxidation process. SO_2 consistently constituted the majority of all sulfurous gases, with only a minimal fraction of H_2S detected. As depicted in Figure 4G, the introduction of CaO facilitated the removal of sulfurous gases from the CLOU process, exhibiting an impressive desulfurization efficiency of 98%.

Flexible fuel adaptability

CLC exhibits exceptional fuel flexibility, enabling the utilization of a wide range of fuels, from gaseous sources (natural gas, refinery gas, or gasification-derived gases) to solid fuels (coal, biomass) and even liquid fuels (heavy oil, bio-oil), which from its flexible reactor and OCs design. This adaptability positions CLC capable of addressing regional energy diversity challenges and integrating renewable or low-grade fuels.

Gaseous fuels. CLC was initially developed for gaseous fuels, which typically exhibit high reactivity and low impurity content. The direct interaction between gaseous fuels and solid metal oxides, coupled with minimal mass transfer limitations, enables high fuel conversion, efficient CO_2 capture, and stable operation in pilot-scale units.¹¹ The true advantage of CLC technology lies in its capability to process more challenging solid and liquid fuels.

Liquid fuels. The use of liquid fuels has recently garnered significant attention. In fact, the conceptual design of CLC reactors for liquid fuels does not differ substantially from those used for gaseous fuels; however, careful consideration must be given to the method of introducing liquid fuels into the reactor. As illustrated in Figure 5A, Moldenhauer et al.^{96–98} employed an injection system in which Sulphur-free kerosene is evaporated, mixed with superheated steam, and directly introduced into a 0.3 kW_{th} CLC unit, achieving continuous operation and high fuel conversion. Furthermore, they observed that the presence of sulfur enhanced the reactivity of ilmenite OC. de Diego et al.^{99–100,101} focused on bioethanol and refinery by-products, employing the same vaporization and premixing method to introduce these fuels into a 1 kW_{th} CLC unit for continuous experiments, achieving near-complete fuel conversion. However, for heavy liquid fuels (e.g., fuel oil and vacuum residue), their high boiling points render evaporation challenging, often leading to pyrolytic carbon deposition. As shown in Figure 5B, Moldenhauer et al.¹⁰² designed a 10 kW_{th} unit for heavy liquid fuels, where the fuel is injected directly into the fluidized bed through a steam-cooled nozzle, preventing pyrolytic carbon deposition prior to evaporation. Using ilmenite and calcium manganite as OCs, they successfully achieved 67 h of continuous operation.

Solid fuels. The CLC process of solid fuels is more complex and can be facilitated through three approaches: syngas CLC, iG-CLC , or CLOU, as previously described. Syngas CLC requires a coal gasification step, which necessitates additional gasifiers and air separation units. Consequently, research efforts have primarily focused on iG-CLC and CLOU. In these processes, solid fuels such as coal, petroleum coke, and biomass are typically introduced into the FR alongside OCs via screw feeders. Coal, the most commonly used solid fuel in CLC, varies significantly in quality depending on its rank. Low-rank coals, such as lignite, exhibit high volatile matter content and reactivity, facilitating easier gasification in iG-CLC .¹⁰³ In contrast, high-rank coals, such as

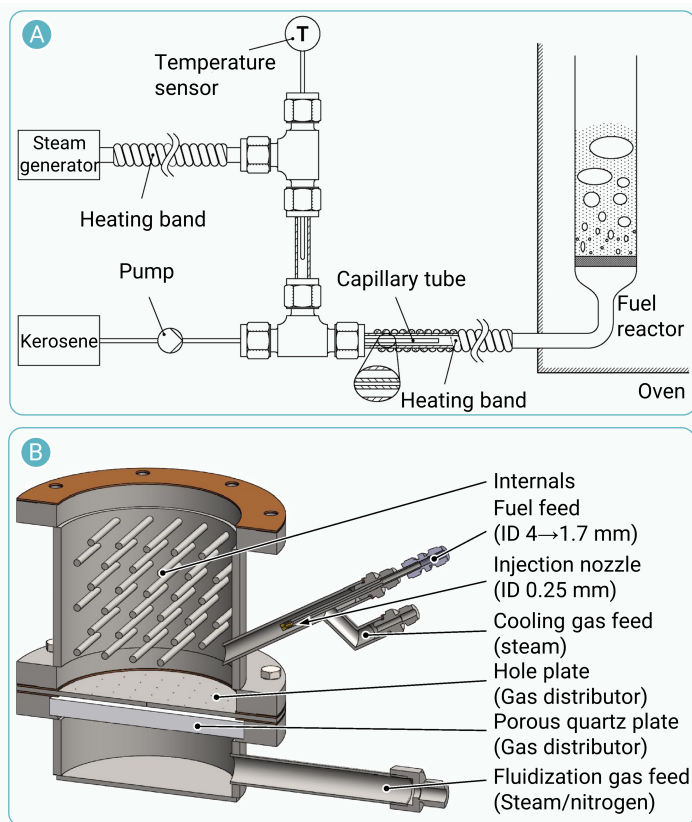


Figure 5. Schematic description of the liquid fuel injector (A) The injection principle for kerosene injection,^{96–98} (B) The direct injection principle for heavy liquid fuels.¹⁰²

anthracite, have lower volatile content and higher fixed carbon, resulting in slower gasification reactions.¹⁰⁴ Nevertheless, due to the greater abundance and higher heating value of high-rank coals, researchers have addressed these challenges by designing appropriate OCs.^{105–106} Furthermore, sulfur impurity poses a challenge when using coal as fuel, which usually contains a high amount of sulfur contaminant (0.3–8%).¹⁰⁷ In CLC, gaseous sulfur compounds can be effectively managed by introducing CaO for in-situ desulfurization, achieving up to 98% sulfur removal efficiency.⁹⁴

Beyond conventional fossil fuels, CLC presents significant opportunities for integration with renewable energy sources. Biofuels, when combined with CO₂ capture and storage technologies (BECCS), provide an effective pathway for achieving negative carbon emissions by removing CO₂ from the atmosphere. Thus, the CLC of biofuels (Bio-CLC) represents a promising BECCS technology.¹⁰⁸ Biogas, a renewable fuel generated through the anaerobic digestion of organic matter, has been utilized in CLC processes, though its application is predominantly in chemical looping reforming processes.^{109–110} Notably, for solid fuels, CLC is often implemented using biomass, which contains significant impurities and high ash content. Conversely, for liquid fuels, CLC can be achieved with highly clean fuels such as bioethanol or biodiesel.¹⁰¹ As these are secondary fuels, their use appears economically unfeasible. Consequently, future research should prioritize the CLC process for biomass.

Although the CLC characteristics of various biomass types have been evaluated in lab-scale reactors and continuous operation units ranging from 0.5 kW_{th} to 4 MW_{th},¹¹¹ the distinct properties of biomass compared to coal introduce new challenges to the CLC process.¹¹² Biomass exhibits a higher content of volatiles (60–85%), which can impede effective contact between OCs and volatiles. This can result in incomplete conversion, necessitating an oxygen polishing step to achieve complete oxidation.¹¹³ Using OCs with oxygen uncoupling properties represents a promising solution. Bio-CLC with CLOU mode has been conducted in continuous units employing OCs based on copper, Cu-Mn oxides,^{114–115} and manganese ore.⁵² These materials enable complete combustion of biomass, yielding a nearly pure CO₂ stream. For the reactor, a multi-stage FR design^{116–117} or the incorporation of a volatiles

distributor¹¹⁸ can be adopted. Additionally, biochar is more reactive than char from fossil fuels, facilitating easier gasification. This enhanced reactivity is attributed to the greater porosity and surface area of biochar, as well as the catalytic effects of higher alkali content, such as K and Na, present in the ash.¹¹⁹ The flexibility of CLC has been extended to the co-combustion of coal and biomass, offering a transitional pathway for industries reliant on fossil fuels. Ohlemüller et al.¹²⁰ conducted 110 h of continuous experiments in a 1 MW_{th} CLC unit, demonstrating that co-firing biomass with hard coal enhances carbon capture efficiency and achieves a CO₂ reduction efficiency of 122%, effectively resulting in negative CO₂ emissions. However, we recognize that integrating biomass into CLC systems presents certain challenges. The variable composition of biomass requires tailored OCs and reactor conditions to ensure efficient combustion and CO₂ capture.¹²¹

UNSOLVED DISADVANTAGES

High sunk cost of OCs

As a core component of CLC, the long-term stability of OCs under cyclic redox conditions is critical for system reliability and operational continuity.¹²² In practical CLC continuous operation, OC particles undergo rapid redox cycles under intense heat and mass transfer conditions while experiencing vigorous fluidization motion. Consequently, strong collisions occur between particles and the reactor. Currently, OCs mainly face the following deactivation mechanisms in the CLC process. Temperature fluctuations exacerbate sintering, particularly for synthetic OCs with high metal oxide content. For instance, Cu-based OCs sinter readily above 900 °C due to low Tamman temperatures, forming dense aggregates that impede gas-solid contact and oxygen release.^{123–124} Besides, Fe segregation and enrichment at the surface can accelerate the thermal sintering of Fe-based OC.¹²⁵ A widely adopted approach involves doping inert supports (e.g., Al₂O₃, TiO₂, ZrO₂) into synthetic OCs to suppress sintering and phase segregation.^{125–127} Mechanical degradation, such as abrasion in fluidized beds, progressively reduces particle size, increasing elutriation losses and operational costs. Previous studies indicate that the attrition rate of OCs is not constant,^{128–129} and their mechanical properties exhibit significant degradation after prolonged operation, making it challenging to accurately predict their service life through direct calculations. Consequently, the actual lifespan of OCs is substantially shorter than their theoretical lifespan. Besides, the sulfur content in the fuel may react with OC to form metal sulfides or sulfates (e.g., Ni, Cu), forming sulfides (Ni₃S₂, Cu₂S) that poison redox activity.^{130–131} Carbon deposition on OC surfaces during incomplete fuel conversion blocks active sites and exacerbates agglomeration.¹³²

Sintering and agglomeration of OCs not only necessitate significant bed material replenishment, thereby increasing operational costs but can also induce severe defluidization of the bed.^{133–134} The majority of currently developed OCs fall significantly short of the theoretical lifespan required for industrial operation.¹³⁵ Standardized testing protocols and real-world performance data are urgently needed to bridge the gap between laboratory evaluations and industrial requirements.

Low combustion and CO₂ capture efficiency

Efforts have been made to achieve high combustion and CO₂ capture efficiency at a cost as low as possible in the κ G-CLC process. The combustion efficiency reflects the degree of conversion of gaseous fuels (or pyrolysis/gasification products in CLC of coal) in the FR, while the CO₂ capture efficiency reflects the degree of conversion of solid fuel (char). However, most continuous CLC units reported in the previous studies were unable to simultaneously achieve both combustion and CO₂ capture efficiency beyond 95%, especially when using high-rank coal as fuel.¹² This suggests that the leakage of fuel in the FR represents a significant limitation for CLC, especially solid fuels.

For gas fuel, the main reason is the limited OC reactivity and inadequate interaction between gaseous fuel and OC particles. In fact, the radial diffusion of gaseous fuel in FR is usually inadequate due to the influence of gas flow entrainment or fuel feeding point layout, resulting in some gas passing through the bed as bubbles, thereby affecting the efficient interaction between fuel and OC. For solid fuels, the chemical reactions between the fuel and the OC in the FR mainly involve heterogeneous reactions between the

reducing gases and the OC. However, solid-solid reactions between char particles and the OC are difficult to occur. The coal devolatilization reaction occurs on a timescale of seconds, while the reaction process for char gasification takes place on a timescale of minutes, with the reaction time ranging from several minutes to tens of minutes depending on the coal rank. Therefore, char gasification is the rate-limiting step in the *i*G-CLC process, and char particles require a longer residence time to achieve higher conversion. Additionally, the hydrodynamic behaviours of char and OCs are different due to variations in particle properties such as density and particle size, thereby hindering complete homogenization. The char particles undergo segregation during the fluidization process and are more likely to exit the FR. As a result, the average residence time of char in the FR is usually shorter than that of OCs. Based on the reasons above, a significant portion of ungasified residual char slips out of the FR and eventually combusts in the AR. The resulting CO₂ mixes with air, thus leading to a lower combustion and CO₂ capture efficiency.

High energy penalties in operation

The CLC process also suffers from the drawback of high operational energy penalties, typically involving coal grinding, fluidization blower, steam fluidization, oxy-polishing and CO₂ purification and compression. In the case study of a 1000 MW CLC-CFB⁵⁷ (Figure 6), the fluidizing blower accounted for a total energy consumption of 8 MW, with 3 MW dedicated to fan heat consumption and 5 MW allocated for steam generation. Nevertheless, a lower air ratio in CLC results in a reduced loss of flue gas in comparison to conventional CFB. In order to enhance the rate of char gasification in the FR, the CLC process necessitates the use of smaller fuel particles compared to conventional fluidized bed combustion of coal. Consequently, grinding the fuel to the required size will consume around 1 MW of heat. Given that the exhaust gas from FR typically retains certain amounts of unconverted fuel gas, an oxygen concentration of 5-15% is necessary for oxy-polishing, resulting in a heat consumption of approximately 5 MW. Moreover, the most significant energy penalty arises from CO₂ purification and compression, which totals approximately 30 MW. Efforts to enhance the efficiency of large-scale CLC units should prioritize the optimization of operating energy penalty. Therefore, future research should focus on integrating and optimizing the CLC process to address this issue.

High process complexity

Furthermore, the process flow of CLC is more intricate. Although achieving excellent CLC performance in laboratory scale devices is possible, the process of scaling up this approach is hindered by challenges of the complex structure and operational procedures of the reactor, in addition to the absence of principles in design and operation.¹³⁶ The CLC process often takes place in interconnected fluidized beds, which is different from the conventional combustion process. As mentioned before, supplementary equipment like a steam generator, char separator, and oxygen polishing are necessary to support the operation of the CLC process.

The interconnection of the FR and AR is influenced by the control of solids circulation and the maintenance of pressure equilibrium, which restricts the adaptability of the system. Generally, a high solid circulation rate is required to guarantee sufficient lattice oxygen supply and efficient heat transfer between the AR and FR. Thus, a substantial inventory of solids is essential to ensure adequate reaction and contact between the OC and fuel. For example, a 300 MW_e coal CLC unit requires a solid inventory and a solid circulation rate of more than 750 t and 5 t/s, respectively, while a conventional CFB requires 300 t and 1-3 t/s, respectively. The elevated levels of solid inventory and circulation rate contribute to increased operational costs and pose challenges in achieving stable operation, leading to issues such as unstable fluidization, empty loop-seal, and inverse gas flow within the loop-seal.¹³⁷ Besides, the OCs will endure intense mechanical, chemical, and thermal stresses during the rapid cycling process, leading to more intensified sintering and agglomeration problems of the particles, which often bring some operational challenges, including de-fluidization, distributor plugging, and ultimately complete collapse of the fluidized bed.

POTENTIAL OPPORTUNITIES

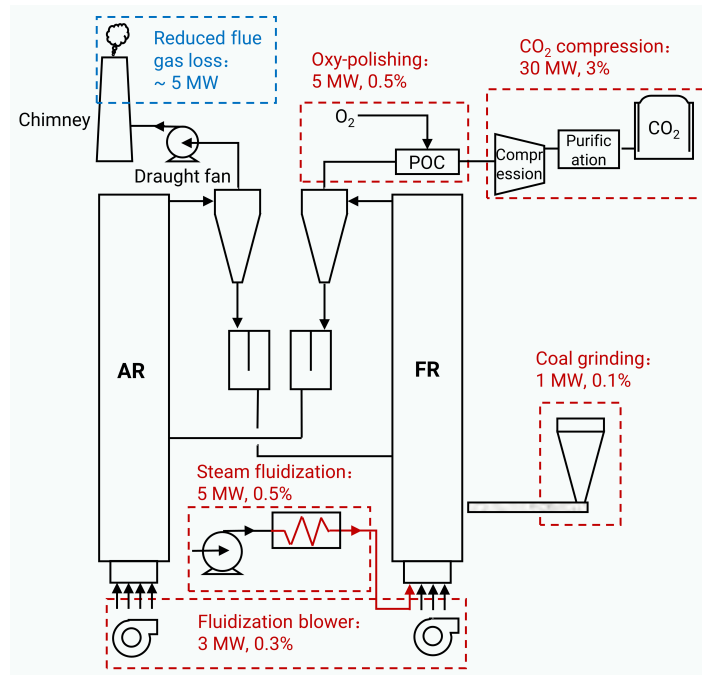


Figure 6. Energy penalty for a 1000 MW_{th} CLC-CFB.⁵⁷

Significant cost advantage

It should be noted that the high cost associated with CO₂ capture has hindered the transition from engineering practices to the commercial implementation of CCUS. In fact, the cost of carbon capture in power plants primarily consists of two factors: the energy penalty associated with CO₂ separation and compression, which results in a loss of power generation efficiency and cost penalties.¹³⁸ The first-generation carbon capture technologies (including post-combustion capture, oxy-fuel combustion, and pre-combustion capture) have already begun to be used in industrial demonstrations in major developed countries around the world. However, due to the additional energy requirements involved in the separation process, power generation efficiency will be decreased by approximately 6-11%, 9-14%, and 7-11%, respectively, for pre-combustion capture, post-combustion capture, and oxy-fuel combustion technologies.¹³⁹ This is economically untenable since it would result in a roughly half-century regression in the efficiency of existing electricity generation.

CLC has the inherent CO₂ separation capability, offering significant advantages in reducing the energy penalty and cost of carbon capture. The CLC technology has been regarded by the U.S. Department of Energy,¹⁴⁰ and the European Union as one of the most promising carbon capture technology.¹⁴¹ Zhang et al.¹⁴² found that the actual thermal efficiency of post-combustion capture, pressurized oxy-fuel combustion, and staged gasification power plants is lower than that of power plants without CO₂ capture due to the energy penalty of capture. In contrast, the performance of CLC power plants is comparable to systems without capture. Ogidiama et al.¹⁴³ analyze the techno-economic performance of a 1000 MW_{th} natural gas-fired power plant utilizing CLC technology. The results show that the plant efficiency and cost of electricity for the CLC plant are 55.6% and 5.5 cents/kWh, respectively, significantly lower than the 50.6% and 6.1 cents/kWh for Natural Gas Combined Cycle (NGCC) power plant with CO₂ capture. The advantages of low energy penalty and cost associated with CLC provide crucial technical support for successfully implementing "carbon peaking and carbon neutrality" goals.

Expected leap in development

Over the past two decades, significant progress has been made in the design, construction, and operation of CLC reactors. Presently, there are more than 40 CLC test units worldwide with varying scales, which range from lab-scale (0.2 kW_{th}) to semi-commercial (4 MW_{th}). In 2001, a test for CLC was conducted initially in a 10kW_{th} CLC unit using natural gas as fuel and NiO as

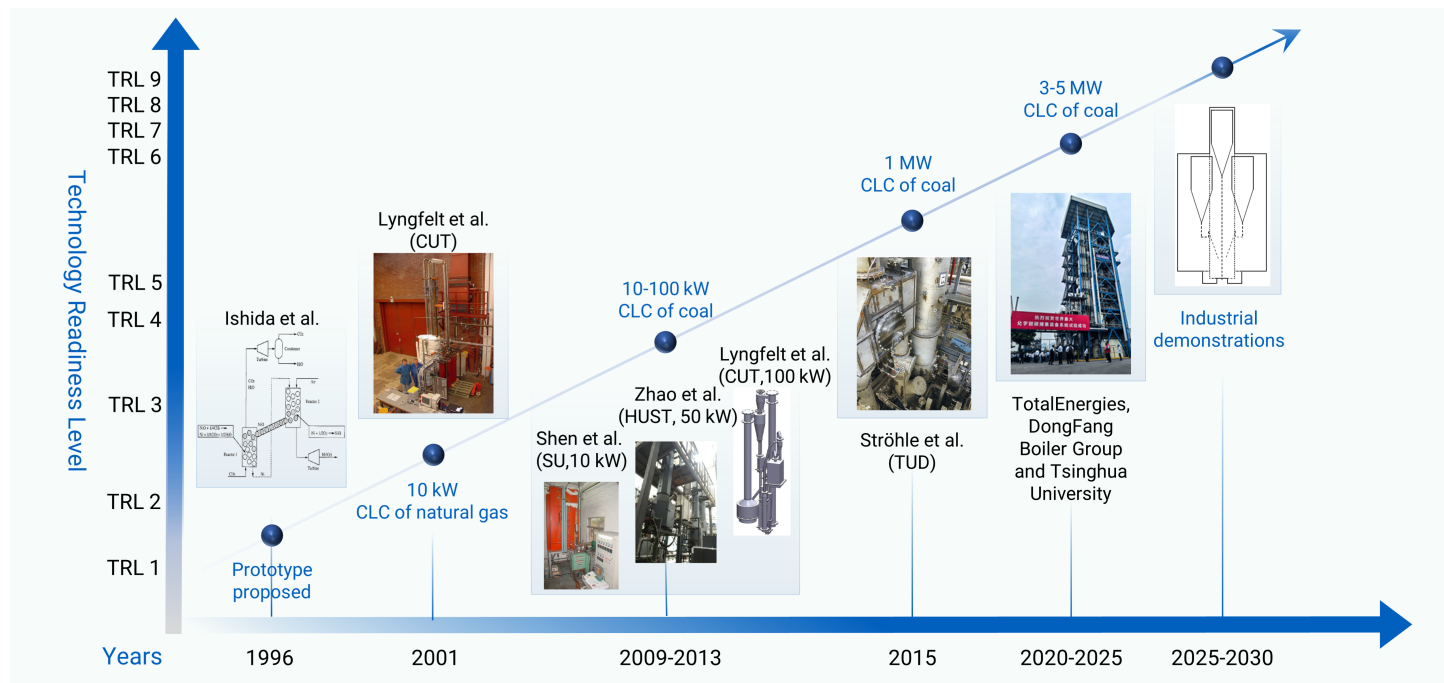


Figure 7. Development of the CLC units.

Table 1. The process of conventional combustion, CLC and CLV.

	Direct combustion	CLC	CLV
Energy conversion	Chemical Energy→Thermal Energy (One-step)	Chemical Energy of Fuels→Chemical Energy of OCs→Thermal Energy (Two-steps)	Low-value Chemical Energy→High-value Chemical Energy (Multi-step)
Conversion steps	$C_nH_m + \text{Air} \rightarrow CO_2 + H_2O$	$C_nH_m + MeO_y \rightarrow MeO_{y-1} + CO_2 + H_2O$ $MeO_{y-1} + \text{Air} \rightarrow MeO_y$	$C_nH_m + MeO_x \rightarrow MeO_{x-2} + C_nH_b + CO_x + H_2$ $MeO_{x-2} + CO_2/H_2O \rightarrow MeO_{x-1} + CO/H_2$ $MeO_{x-1} + \text{Air} \rightarrow MeO_x$

OC for 100 h total continuous operation with a fuel conversion efficiency of 99.5%.¹⁴⁴ Since then, numerous long-term tests utilizing gaseous fuels have been performed, such as those from CUT in Sweden,¹⁴⁵ the Korea Institute of Energy Research (KIER),¹⁴⁶ the Spanish Coal Research Institute (ICB-CSIC),¹⁴⁷ Vienna University of Technology (TUV),¹⁴⁸ and the Norwegian University of Science and Technology (NTNU),¹⁴⁹ successively established gas fuel CLC pilot plants with capacities ranging from 0.2 kW_{th} to 1 MW_{th}. It was not until 2008, when CUT achieved the CLC process of coal and biomass char in a 10 kW_{th} reactor,¹⁵⁰ that the practical viability of solid fuel in the CLC process was confirmed. Typical CLC reactors of solid fuel include the 100 kW_{th} coal CLC unit established by CUT,¹⁵¹ the 10 kW_{th} coal/biomass CLC experimental platform,¹⁵² and the 50 kW_{th} pressurized CLC unit¹⁵³ built by Southeast University (SU), the 50 kW_{th} coal-derived CLC unit set by the ICB-CSIC¹⁵⁴, the 25 kW_{th} dual fluidized bed reactor established by Hamburg University of Technology (HUT)¹⁵⁵, and the 50 kW_{th} CLC reactor constructed by Huazhong University of Science and Technology (HUST),¹⁵⁶ etc.

Among them, Darmstadt University of Technology (TUD),¹⁵⁷ Alstom Power,¹⁵⁸ and CUT¹²¹ have successfully completed pilot tests at the MW-scale. Besides, two demonstration units of 3 MW_{th} in Ningxia University,¹⁵⁹ China and Ulsan, South Korea¹⁶⁰ are under construction. Recently, the world's largest CLC unit with a thermal power of 5 MW_{th} was completed in Deyang City, outside Chengdu in China,¹⁶¹ which achieved full autothermal CLC mode with the lignite/petcoke fuel mixture and maintained around 60 h. The operating results show that the CO₂ capture efficiency of CLC reached 95% for lignite and 75% for petroleum coke, with the dry-basis CO₂ content at the outlet of the FR exceeding 93% and oxygen demand less than 10%.¹⁶² This indicates that the CLC unit might be used as a stand-alone facility to capture CO₂, which accounts for technology readiness level (TRL) = 6¹⁶³ (Figure 7). Those remarkable advancement will further drive CLC technology toward industrial application, with expectations of gradually progressing to industrial demonstration in the near future. As more and more scholars dedicate their

efforts to CLC technology, it will have more new applications and expansions, which will undoubtedly be a significant advantage for the development of this field.

Chemical looping beyond combustion

Recent developments in chemical looping (CL) research reveal that the application of this concept has extended far beyond its traditional use in CLC. With deeper exploration and utilization of the CL concept, a broader definition of CL processes can be proposed: decomposing a chemical reaction into two or more reactions occurring in different spaces or at different times, utilizing a recyclable medium to transfer substances and energy, while simultaneously achieving *in-situ* separation of reaction products.¹³ The chemical looping valorization (CLV) process maximizes the potential of chemical energy by employing chemical looping partial oxidation (CLPO) to convert diverse fuel sources into specific chemicals, rather than fully oxidizing them and converting the chemical energy into thermal energy, as is done in the CLC process. Table 1 outlines the main features of conventional combustion, CLC and CLV.

The processes of CLV are always related to the directed transformation of reactant molecules. Selective oxidation of light alkanes, aromatic hydrocarbons, and biomass to desired products can be achieved by tailoring the active sites on the OCs, the lattice oxygen-carrying medium, and the rate of lattice oxygen migration. The earliest application of CLPO can be traced back to DuPont's proposal for the selective oxidation of butane, achieving a selectivity higher than 90% towards maleic anhydride.¹⁶⁴ In recent years, the applications of CLPO have gradually expanded its applications in various fields. Due to the rapid growth of available methane, ethane, and propane in shale gas production, the CL conversion of light alkanes has gained increasing attention. In 2016, the research group of Professor Li at North Carolina State University proposed a two-step redox-based process for ethane chemical looping oxidative dehydrogenation (CL-ODH) under an O₂-free environment.¹⁶⁵ The absence of gaseous oxygen in this process effectively

inhibits the nonselective reaction routes and diminishes the risk of explosion, leading to an 82% reduction in CO₂ emissions in comparison to the steam cracking process. Additionally, the chemical looping reforming of methane,¹⁶⁶ chemical looping oxidative coupling of methane,¹⁶⁷ and chemical looping dehydroaromatization¹⁶⁸ have also been the focus of recent research. Other novel effective processes include chemical looping epoxidation of ethylene developed by Chan et al. at the University of Cambridge,¹⁶⁹ chemical looping selective oxidation of H₂S to elemental sulfur reported by Kane et al.,¹⁷⁰ and chemical looping ammonia oxidation for NO production proposed by the research team from Dalian Institute of Chemical Physics.¹⁷¹

If the solid carrier is a nitrogen carrier, CL technology can be applied in ammonia synthesis. The CL-ammonia synthesis process decouples the catalytic process of ammonia synthesis into N₂ fixation and hydrolysis reactions through the circulation of nitrogen carriers. Compared to the conventional Haber-Bosch process, which necessitates tough temperature and pressure (350–550 °C and 15–25 MPa) and a large amount of energy,¹⁷² this process can be conducted under milder conditions (atmospheric pressure, 100–300 °C), thereby avoiding the gas compression process with high energy penalty and significantly improving the single-pass yield.¹⁷³

From an overall viewpoint, the idea of CL can be implemented in any catalytic process if suitable carriers capable of transferring desired functional groups can be discovered. The inherent benefit of segregating reactants and products during the CL process has the potential to significantly impact various chemical procedures in the future, resulting in improved efficiency and more environmentally friendly chemical processes.

REMAINING CHALLENGES

Trade-off between OC performance and cost

While advancing CLC technology, the large-scale preparation of high-performance OCs has consistently posed one of the most critical challenges to be addressed. As previously mentioned, although synthetic OCs exhibit excellent performance in CLC, they suffer from high initial costs, as well as high sunk costs caused by particle abrasion, sintering, and poisoning. In contrast, natural ores and industrial waste materials are cheap and have excellent resistance to sintering and agglomeration, but their application is limited by their relatively low reactivity. Figure 8A roughly illustrates the relationship between the reaction performance and cost of several representative OCs.¹⁵⁹ In general, highly reactive OCs tend to be associated with higher costs. In this case, striking an optimal equilibrium between the performance and cost of OCs is vital.

The poor reactivity of OCs originating from natural ores and industrial waste can be improved by mixing different low-cost materials to achieve a synergistic effect. Yang et al.¹⁷⁴ simply mechanically mixed copper ore with iron ore and the prepared composite OCs could show synergistic effects in improving the oxygen transport capacity and reaction rate of iron ore and the anti-agglomeration properties of copper ore. The cost of the OC is \$0.72/kg. Due to the distinct physical properties between different particles and the poor homogeneity of the samples prepared by mechanical mixing, segregation phenomena are prone to occur in the fluidized bed reactor. Thus, the pilot-scale production of composite OCs was successfully achieved using spray drying,¹⁷⁵ extrusion spherulization^{176–177} and hydroforming¹⁷⁸ methods. Among them, Wu et al.¹⁷⁷ used red mud, copper ore, and montmorillonite as raw materials to prepare an industrial-scale bimetallic OC using the extrusion spherulization method, and the raw material cost of the OC is only \$0.27/kg. In a batch fluidized bed reactor, they achieved a CO₂ gas production of over 94.5% and a combustion efficiency exceeding 94.7% (Figure 8B).

For synthetic OCs, perovskite structures exhibit several valuable characteristics, including reduced raw material costs, more environmentally friendly materials, and the ability to release oxygen from their structure. Particularly, Ca-Mn perovskite has been identified as a promising material for scale-up.^{179–181} Liu et al.¹⁸² prepared CaMn_{0.5}Ti_{0.37}Fe_{0.125}O_{3-δ} (CMTF_Ind) OC at the industrial level via spray drying granulation. As shown in Figure 8C, CMTF performs similarly to its laboratory counterpart (CMTF_Lab), showing high stability over multiple redox cycles without degradation or agglomeration. Additionally, the CMTF_Ind particles require significantly lower solid inventory and fan energy consumption in CLC reactors compared to other OCs, which was once considered for potential deployment in a 3 MW CLC pilot plant.¹⁶¹ However,

the CMTF OC suffers agglomeration in a fluidized bed thermogravimetric analyzer, indicating it may have thermal stability issues.¹⁸³ Wu et al.¹⁸⁴ synthesized a novel perovskite CaMn_{0.625}Ti_{0.125}Fe_{0.125}Mg_{0.125}O_{3-δ} (CM_{0.625}TF-Mg) by substituting the B-site with Mg via hydroforming method, where Mg substitution eliminates the activation effect and promotes lattice oxygen release (Figure 8D). CM_{0.625}TF-Mg not only possesses a high oxygen donation ratio (~5.0 wt.%) but also demonstrates excellent thermal and chemical stability during multiple redox cycles, with a service life of 8,333 h in attrition test. Its material cost (\$0.892/kg) and usage cost (\$0.00217/kg[O]/h) are notably low, making it highly economical for industrial applications.

With the advancement of high-throughput computing and artificial intelligence (AI) algorithms, the tailored-design of OCs can be systematically optimized by leveraging automated or parallelized techniques to modify design parameters. This approach effectively reduces the time and cost associated with the traditional “trial-and-error” experimental methodology. By integrating AI-driven combinatorial screening of OCs with limited experimental evaluation, an efficient and rapid workflow for exploring the design space can be established. Yan et al.¹⁸⁵ proposed a methodology utilizing machine learning (ML) algorithms to predict the reactivity of manganese ores as OCs in chemical looping processes. The basic workflow, as illustrated in Figure 8E, includes the following steps: 1) defining the target problem and process; 2) collecting training data to build a dataset; 3) training and optimizing the ML model; and 4) using the optimized model to predict the performance of previously untested OCs. The results demonstrated that a trained artificial neural network (ANN) model can provide highly accurate performance predictions for heterogeneous and multi-component OC. In the future, coupling ML algorithms with high-throughput performance prediction and screening methods for OCs will be of great significance for optimizing the balance between cost and performance of OCs, as well as promoting their large-scale preparation and application.

Management of heat and mass transfer and rate-limiting reaction

On the one hand, based on the fundamental principles of CLC, the endothermic demand of gasification reactions in the FR can be supplied by the sensible heat of oxidized OCs from the AR. Theoretically, the CLC system can achieve autothermal operation without external heat input. The sensible heat of the OCs is primarily regulated by the solid circulation rate. Therefore, for large-scale interconnected fluidized bed CLC reactors, a very high solid circulation rate is typically required. On the other hand, the optimal temperature for achieving rapid gasification of coal char is generally above 1300 °C, whereas the operating temperature range for CLC is typically between 800–1000 °C.¹⁸⁶ As a result, the gasification rate of coal char within this range is more than an order of magnitude slower than the reduction/oxidation rates of the OCs (Figure 9A).

In summary, the substantial heat generated by the rapid oxidation reactions in the AR needs to be effectively and instantaneously transferred to the FR, which requires a high solid circulation rate. However, in the FR, a relatively slower solid circulation rate is more conducive to ensuring sufficient contact and reaction between the OCs and the solid fuel, thereby achieving a higher fuel conversion rate. Therefore, effective management of heat and mass transfer, along with the regulation of slow reactions, becomes critical to the design and operation of the reactor.

Attain effective management of heat and mass transfer. The regulation of heat and mass transfer is a critical aspect of optimizing the operation of CLC reactors. To facilitate the flexible circulation of bed materials, enhance gas-solid contact, and promote fuel conversion, it is essential to implement targeted measures. This can generally be achieved from two perspectives: OCs and the reactor itself. The former primarily involves the regulation of the reaction heat of the OC (i.e., thermoneutral OCs), while the latter focuses on the regulation of the circulation rate of the OCs.

To reduce the required OC circulation rate, thereby minimizing regulation complexity and operational costs, an effective approach is to employ OCs that enable the FR to exhibit overall thermoneutral or slightly exothermic behaviour. This can be achieved by regulating the reaction heat of the oxygen carriers during oxidation and reduction processes. Yang et al.¹⁷⁴ discovered that there are synergistic effects between iron ore and copper ore. As shown in Figure 9B, copper ore releases heat with syngas in the reduction stage,

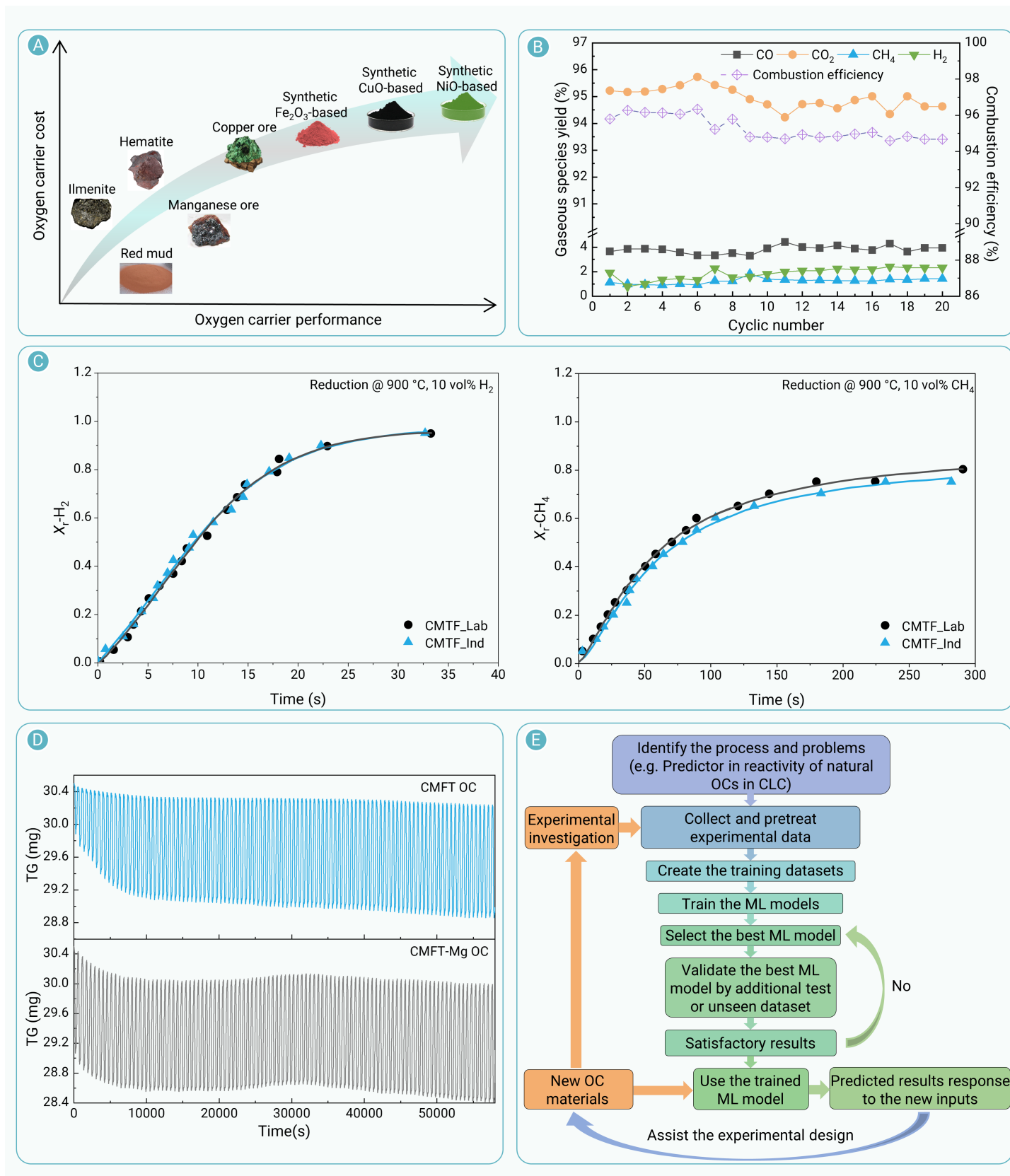


Figure 8. (A) Correlation between the performance and cost of typical OCs;¹⁵⁹ (B) Gaseous species yields and combustion efficiency among 20 cycles at 950 °C;¹⁷⁷ (C) Comparison of the heterogeneous reduction kinetics between CMFT_Ind and CMFT_Lab;¹⁸² (D) TG curves of CMFT and CM_{0.625}TF-Mg OC;¹⁸⁴ (E) Workflow of developing a ML model for OCs in the CL process.¹⁸⁵

whereas hematite absorbs heat. When the mixing ratio of copper ore is 20 wt%, it exhibits an initial exothermic reaction followed by an endothermic one. Thus, the whole reduction stage is approximately thermally neutral, which

helps to regulate the temperature of the FR.

Besides, it is possible to regulate the circulation rate by adjusting the gas flow rates in different parts of the reactor system, thereby influencing the

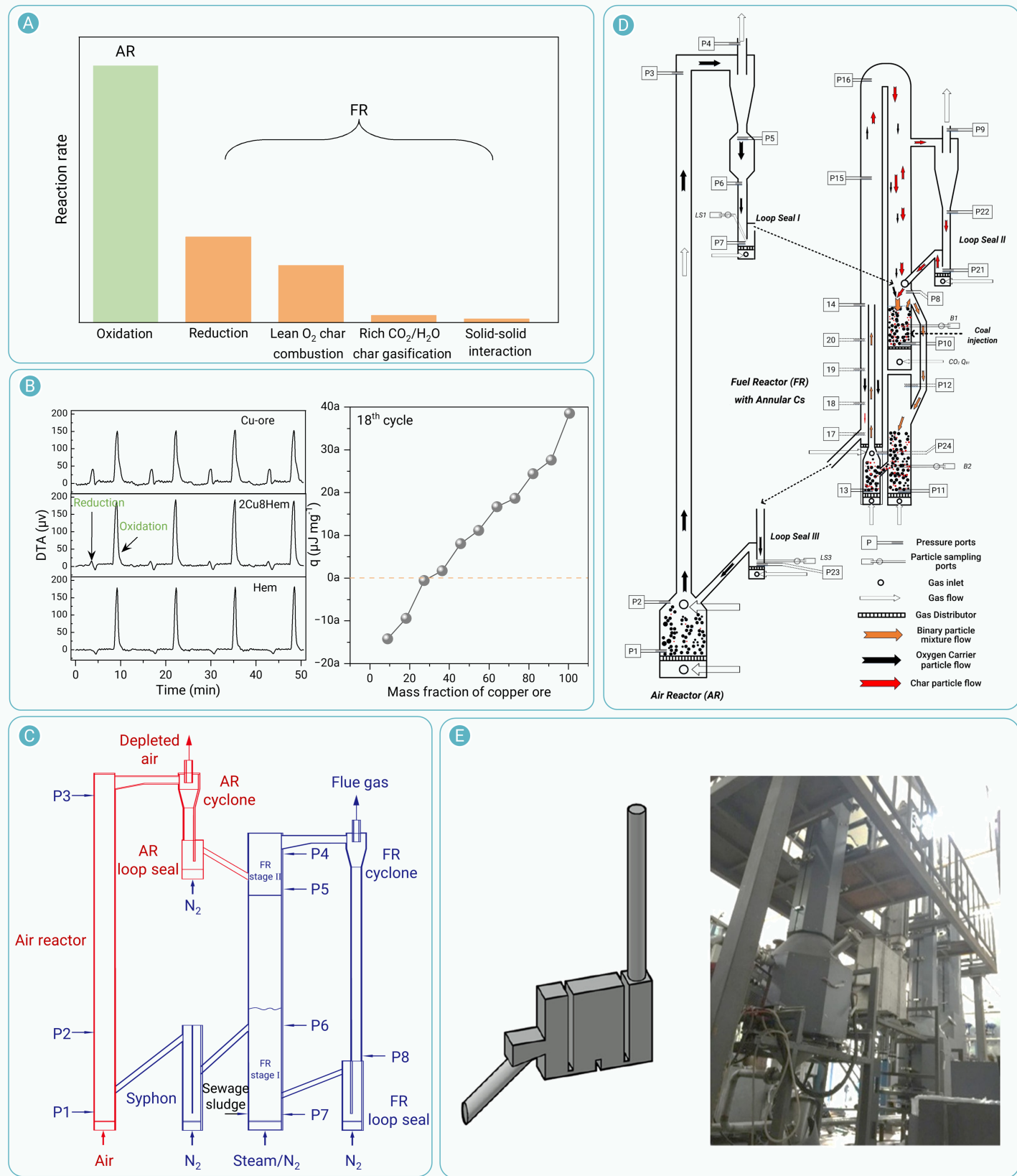


Figure 9. (A) Relative magnitudes of reaction rates of various reactions in CLC/CLOU of coal;¹⁵⁹ (B) The DTA curves and heat release capacity of Copper-iron ore OCs;¹⁷⁴ (C) 5 kW_{th} CLC reactor at SU;¹⁸⁹ (D) 30 kW_{th} CLC reactor at Tsinghua University;¹⁹⁰ (E) Schematic diagram of CS and a picture of the 50 kW_{th} iG-CLC reactor.¹⁵⁶

temperatures in the AR and FR as well as the reactions occurring within them. Simply adjusting the gas flow rates in the AR and FR often leads to adverse changes in the operational state of the reactors. To address this issue, cyclones equipped with loop seals (LS) or bidirectional loop seals can

be used to establish an internal circulation bypass for bed materials,^{154,187–188} which enables partially returning ejected bed material from AR/FR back to the same reactor and partially to FR/AR. By adjusting the internal circulation ratio, this approach allows precise control of the solid

circulation rate between the AR and FR.

Facilitate the rate-limiting step. As previously mentioned, the gasification of coal char is identified as the step that limits the rate of the coal $\dot{g}$ -CLC process. Thus, it is a pivotal concern in $\dot{g}$ -CLC to control and enhance the conversion of coal char, which can be achieved specifically by extending the reaction time of the char and enhancing its reaction rate.

Prolonging the residence time of char particles in the FR is an effective strategy to enhance their gasification. This can be achieved by optimizing the reactor structures, including modifications to the FR (such as multi-stage FR and internal solid circulation in the FR) and the integration of CS. For the former, typical systems include the 5 kW_{th} reactor at SU¹⁸⁹ (Figure 9C) and the 30 kW_{th} reactor at the Tsinghua University¹⁹⁰ (Figure 9D). As for the latter, ungasified char particles in the FR can be separated from the OC and sent back into the FR for further gasification using CS, or gasified and reintroduced the gaseous products back into FR using gasification carbon stripper (GCS). Most approaches demonstrate strong performance in practical applications, achieving separation efficiencies above 80% and carbon capture efficiencies exceeding 90%.¹⁹¹ For example, a four-chamber LS positioned in a 50 kW_{th} CLC reactor of HUST, as shown in Figure 9E,¹⁵⁶ integrates a two-chamber GCS between the supply chamber and the recycle chamber of the LS. This configuration achieved a maximum CO₂ yield of 91% and a combustion efficiency of 86% at 1000 °C. Nonetheless, the design and operation of CS remain a significant challenge in operating pilot-scale CLC units, considering the elevated expenses involved and the lack of established design principles.

The process of CLOU³⁴ can be used to avoid slow gasification in $\dot{g}$ -CLC. Zheng et al.¹⁹² suggested that copper-based OCs, which release gaseous O₂ and exhibit CLOU effects, can increase the conversion rate of solid fuels by several times or even tens of times. Even under oxygen-lean conditions, the direct reaction between coal char and O₂ can promote its conversion.¹⁹³ Therefore, employing OC that can emit gaseous oxygen, including copper ore, manganese ore, and Cu-decorated iron ore, may considerably boost the conversion rate of coal char in the CLC process. Moreover, alkali/alkaline earth metal salts have been proven to catalyze the gasification reaction of coal char.^{50,194} Alkali metals (e.g., Na, K) can partially bond with carbon, causing surface charge migration and altering the electronic cloud distribution of carbon atoms on the char surface. This change creates a more chemically reactive structure on the char surface, thereby enhancing the reaction. Alkaline earth metal salts (e.g., Ca, Ba) participate directly in the coal char gasification reaction as catalysts, which can be described through redox reactions.

Development of reactor design and operation methods

The design theory and operational methods of CLC reactors form the foundation of CLC applications, particularly for costly pilot demonstration and industrial-scale systems, a comprehensive design theory becomes especially critical. Although institutions such as TUD,¹⁵⁷ Alstom Power,¹⁵⁸ CUT,¹⁵⁹ and TotalEnergies & Dongfang Boiler Group¹⁶¹ have established MW-scale dual-fluidized bed pilot systems and conducted a series of operational tests, the design process of these reactors has not been publicly reported. The design methods for CLC reactors differ fundamentally from those of traditional combustion process reactors, resulting in a severe lack of corresponding design and operational experience. Moreover, the complex and interrelated gas-solid reaction, flow, heat transfer, and mass transfer processes in the reactor pose significant challenges, making existing semi-empirical reactor design methods inadequate for the engineering design and scale-up of CLC devices. Therefore, efficiently and reliably modelling the "transport-reaction" phenomena within CLC reactors is essential to establishing the theoretical foundation for CLC reactor design methods.

To improve the quality of reactor engineering design, numerical simulation methods, such as the macroscopic simulation and the computational fluid dynamics (CFD) simulation, have been integrated into the design process. The macroscopic model developed by Abad et al.¹⁹⁵ has been used to simulate the performance of CLC pilot plants up to the scale of 1 MW_{th} fluidized bed and has been applied in the numerical design of reactors at various scales.^{196–197} However, macroscopic simulations are unable to capture the details of complex turbulence, gas-solid conversion, and various fluidization states present in the reactors. CFD simulations can accurately model the

evolution of gas-solid reactive flows in the reactors. They not only enable the study of macroscopic heat transfer phenomena in the CLC process^{198–199} but also help optimize the operational and structural parameters of the reactor.^{200–202} This facilitates the design of novel reactors or those with new structures in the absence of engineering experience. However, CFD simulations require high computational costs. Chen et al. from HUST systematically combined traditional reactor engineering design methods with one-dimensional reduced-order simulation methods and three-dimensional CFD simulation methods, proposing a multi-step numerical design approach for reactors. They applied this approach to the design of a 5 MW_{th} coal $\dot{g}$ -CLC reactor^{203–204} and a 1.5 MW parallel packed bed reactor^{205–206} based on engineering design combined with one-dimensional reduced-order simulations, as well as the design of a 10 MW_{th} natural gas CLC reactor^{207–208} based on engineering design combined with CFD simulations.

SUMMARY AND OUTLOOK

As a cost-effective carbon capture technology, CLC will provide an innovative solution for deep decarbonization in global industries such as power generation, heating, petrochemicals, chemicals, and oil and gas. Its inherent high-efficiency carbon capture capability eliminates the need for additional separation equipment, significantly reducing energy consumption and costs, while providing high-purity carbon dioxide for resource utilization. However, as a cutting-edge technology that has only recently entered the engineering demonstration stage, CLC still faces some development bottlenecks.

(1) From both efficiency and economic perspectives, traditional methods for screening OCs are far from sufficient to meet current demands. By leveraging density functional theory combined with transition-state theory, the reaction processes on gas-solid surfaces, internal structural changes of materials, and energy variations in each stage can be described and analyzed at the atomic/molecular level. This approach allows for the efficient, bottom-up preparation of OCs with ideal physical and chemical properties based on both kinetic and thermodynamic principles. Besides, as efficient tools for rapidly predicting material properties, ML methods facilitate the effortless gathering of extensive material information and promote a balance between efficiency and expense by employing automated or parallelized approaches for OC property prediction and extensive screening.

(2) Techniques such as spray drying, cement bonding, hydroforming, and extrusion-spheronization have been developed to combine particles efficiently to produce affordable materials on a large scale. However, powdered OCs often exhibit altered reaction kinetics after the shaping process, which must be reassessed under practical conditions. In addition, although many researchers are dedicated to developing OCs, variations in testing platforms and operating conditions make it challenging to evaluate the prepared OCs on a standardized level. Therefore, we firmly believe that establishing a standardized performance evaluation criterion for OCs is crucial in evaluating their practical utility in real CLC processes based on test results.

(3) The critical challenges in CLC reactor design lie in enhancing gas diffusion conditions and mitigating the impact of heat and mass transfer on gas-solid reactions. Due to the lack of engineering design experience, numerical simulation can provide effective guidance. However, since each method is based on different principles, their complexity and accuracy vary. An efficient and reliable design needs to strike a balance among these factors. First, it should exhibit a high degree of internal consistency. Second, the design methodology should integrate fundamental modules—mass and energy conservation, gas-solid flow, gas-solid conversion, as well as heat and mass transfer—in a coherent and logical manner. Furthermore, various reactor design methods should be employed in an integrated design process, starting with low-precision methods for extensive parameter exploration. The preferred design scheme can then be fed into high-precision methods for validation, optimization, or iteration. Lastly, this iterative approach combines low- and high-precision methods to ensure the design process is both comprehensive and accurate.

(4) When CLC transitions to pilot plants, the system becomes more complex and presents operational challenges. Current demonstration units primarily focus on realizing autothermal operation while neglecting to obtain high combustion and CO₂ capture efficiency simultaneously. In the future, comprehensive experimental research should be carried out on MW-scale

systems, with a stronger focus on compatibility with multiple functionalities, such as adaptability to different types of OCs and fuels. Meanwhile, operating conditions and control strategies should be further optimized to achieve the optimal cost-efficiency balance among OCs, fuels, and reactors.

The “double carbon” goals of carbon peak and carbon neutrality will promote energy transformation in China, making low-carbon emissions the central focus of energy development in the 14th Five-Year Plan and even in a longer period. CLC technology presents unprecedented opportunities for development in China. Therefore, despite certain obstacles to its progress, we firmly believe that through information sharing and collaboration among research institutions, government bodies, and investors, the industrial application of CLC technology can be expected in the near future.

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DECLARATION OF INTERESTS

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

DATA AND CODE AVAILABILITY

No data was used for the research described in the article.