Advantages, Disadvantages, Opportunities and Challenges of Chemical Looping Combustion

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ABSTRACT

Chemical looping combustion (CLC), as a new generation of combustion technology, has the advantages of inherent CO_2 separation and cascade energy utilization. In this paper, the basic principles and development history of CLC are presented, and the inherent technical advantages, potential disadvantages, development opportunities, and key technical challenges are systematically analyzed. Finally, this paper provides an outlook on the technological maturity and industrial application of CLC.

Keywords: CCUS; Chemical looping combustion; Oxygen carriers; Reactor; CO_2 capture

1. INTRODUCTION

CO_2 emissions from the combustion of fossil is the main cause of global warming. However, the global energy supply will still depend on fossil fuels in the long term[1]. Carbon capture, utilization, and storage (CCUS) technology is considered an essential option to achieve deep emission reductions in traditional fossil energy industries. China is the largest carbon emissions country, and its energy consumption still mainly depends on coal [2]. CCUS gives a good solution because it can control CO_2 emissions and support China's coal-dominated energy structure. However, the high cost of conventional carbon capture technologies has hindered the deployment of CCUS.

Chemical looping combustion (CLC), as a promising combustion technique for fossil fuels, can enable in-situ carbon capture during the combustion process, thus having the potential for uniquely low costs of CO_2 capture. Nonetheless, research is still needed to bring the technology to a fully commercial level. This review analyzes the inherent advantages, potential disadvantages, development opportunities, and key

challenges of CLC technology from the perspective of its basic principles, and aims to provide some theoretical guidance for its commercial implementation.

2. CHEMICAL LOOPING COMBUSTION

CLC technology is to decouple the co-feeding process of oxygen and fuel into two independent redox processes. As shown in Fig.1, in the fuel reactor (FR), the active component of the oxidized oxygen carrier (OC) reacts with the fuel, transferring lattice oxygen to the fuel for combustion. As the effluent from the fully combusted FR contains only CO_2 and H_2O, there is no need for additional separation equipment, and high-purity CO_2 flow can be obtained after the condensation of steam. Subsequently, the reduced OC enters the air reactor (AR), regenerating to its oxidized state by air oxidation and releasing heat. Then, OC is returned to the FR for the next redox cycle.
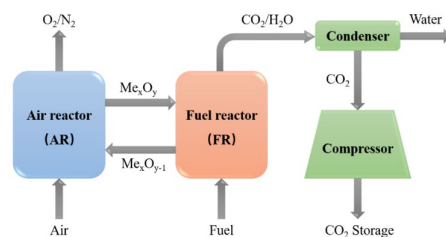
Fig. 1. Schematic diagram of chemical looping combustion

3. INHERENT ADVANTAGES

3.1 Source Enrichment of CO_2

Because fuel can be oxidized without mixing air and the combustion products in the CLC process, the flue gases would be free of N_2, consisting mainly of carbon dioxide and water vapor. After condensation of steam, essentially pure CO_2 is obtained, CO_2 capture is inherent in the process and no gas separation equipment is

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needed. Compared to other CO_2 capture technologies, CLC technology ensures efficient fuel combustion and achieves inherent separation of CO_2 from flue gas, enabling the *in situ* capture of high-concentration CO_2 during the combustion process and eliminating the need for a gas separation step.

The cost of a 1000 MW_{th} boiler for CLC of solid fuels was investigated by Lyngfelt et al. [3]. The total cost of CO_2 capture was estimated to be 22 \$/tonne CO_2 avoided and within the range of 18-29 \$/tonne CO_2, which is much lower than the costs of pre-combustion capture (28-41 \$/tonne CO_2), oxy-fuel combustion (36-67 \$/tonne CO_2), and post-combustion capture (36-53 \$/tonne CO_2) [4]. Besides, they thought a CO_2 capture of 98% could be reached for the 1000 MW_{th} unit without an actual carbon stripper, with only 3.9% energy consumption. Therefore, CLC technology can avoid separation costs with less energy penalty with its ability to source enrichment of CO_2, thus having a better development prospect for CO_2 reduction.

3.2 ENERGY CASCADE UTILIZATION

The direct combustion of fuel leads to the degradation of its high-quality chemical energy into thermal energy, resulting in a larger irreversible loss. In contrast, the energy released in the CLC process is facilitated by the cycling of OCs, which decouples the tough release process of chemical energy into physical energy during direct combustion into the ordered release process of fuel chemical energy, which means from fuel chemical energy to OC chemical energy and finally to thermal energy. This novel approach to energy conversion enables the efficient utilization of chemical energy before fuel combustion compared with direct combustion, thereby reducing the irreversible loss in the chemical energy of fuel converted and degraded into thermal energy. In other words, CLC process can achieve the cascade utilization of chemical energy of fuel in the CLC process [5].

Specifically, there is a large level difference (A_{ch1}-A_{Th}) between chemical energy and physical energy in the conventional combustion process (as shown in Fig.2). However, the CLC process first integrated with the process of chemical reaction, which enables the stepwise utilization of chemical energy of fuel from energy level A_{ch1} to A_{ch2}. Thus, the energy-level difference between chemical and physical energy in CLC combustion is (A_{ch2}−A_{Th}), resulting in less exergy destruction. Zhao et al. [6] found that due to the higher exergy efficiency (71.1%) and lower irreversible loss of the CLC process, the net power generation efficiency of CLC coal-fired power plants was improved by 2.4% compared to that of a plant-based on monoethanolamine with the same carbon capture efficiency. Yan et al. [7] conducted a comparative analysis of direct combustion and CLC, which showed that CLC could reduce the irreversible loss by 10.35% and increase the released heat energy by 6.52% compared to direct combustion. Therefore, CLC can achieve higher thermal and exergy efficiency than traditional combustion methods due to its cascade energy utilization.
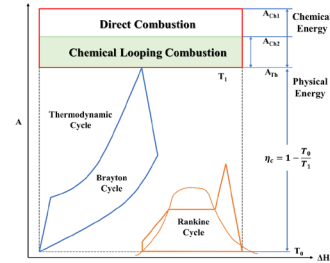

Fig. 2. The concept of the cascade utilization of energy

3.3 In-situ Pollution Control

The combustion and conversion processes of coal are accompanied by the migration and release of pollutants, and the mechanisms of pollutant generation and inhibition have been a research focus. According to previous studies, based on the inherent characteristics of CLC, such as the O_2-free atmosphere, high concentration CO_2/H_2O, and relatively low temperature, pollutants such as dioxins, sulfur, nitrogen, and mercury can be *in situ* inhibited from generation and conversion.

Firstly, because CLC provides an O_2-free combustion pattern, the oxidative cleavage of large carbon molecules and the conversion from HCl to Cl_2 is significantly reduced, inhibiting the low temperature de novo synthesis and precursors reaction of PCDD/Fs. Secondly, CLC can promote the conversion of fuel NO_x to N_2, and it can *in-situ* inhibit the generation of NO_x, allowing meeting the national standard for NO_x emissions without denitrification equipment at the FR. Furthermore, for mercury, Hg^0 is converted to Hg^p and Hg^{2+} through adsorption and oxidation of OC and fly ash, which can reduce the difficulty of downstream treatment. As for sulfur, one idea is to convert it into SO_2 in the FR and then treat the flue gas, and another idea is to convert S to sulfide/sulfate and then solidify it in OC or ash.

4. UNSOLVED DISADVANTAGES

4.1 Low Combustion and CO₂ Capture Efficiency

Most of the reported continuous CLC units in the literature could not attain both combustion efficiency

and CO_2 capture efficiency higher than 95% at the same time [8], indicating that the fuel slip in the FR is still a major disadvantage of CLC.

The slip of gaseous fuels is mainly attributed to the low reactivity of OC and insufficient contact between gaseous fuel and OC particles. To address this issue, researchers have adopted multiple coal-feeding points and multi-stage FR to enhance the interaction of OC with volatiles and char gasification products. However, these must still be verified and optimized in pilot-scale operation CLC unit. As for the slip of solid fuels, the main reason is the severe mismatch of reaction rates in CLC of coal. At present, adding carbon stripper (CS) downstream of the FR is the primary solution to the problem, which can separate the unconverted char particles from the OC particles downstream of the FR and return the char particles to the FR for further conversion. However, due to the limitation of high costs and the lack of design guidelines, the design of CS is still a vital issue to be solved in the operation process of pilot-scale CLC units.

4.2 High Operational Energy Penalties

Another disadvantage of CLC is its relatively high operating energy penalties, which usually include coal grinding, fluidization blower, steam fluidization, coal grinding, oxy-polishing and CO_2 purification and compression.

Take a 1000 MW_{th} CLC-CFB as an example [9], the total energy penalty of fluidizing blower is 8 MW, including 3 MW for fan energy penalty and 5 MW for steam production. However, the low air ratio for CLC reduces its flue gas loss compared with the traditional CFB. Meanwhile, to promote the char gasification rate in the FR, the CLC process requires a smaller fuel size than normal fluidized bed combustion of coal so that the fuel grinding will need about 1 MW of energy penalty. In addition, since the FR exhaust gas usually contains some unconverted fuel gas, 5-15% of O_2 is needed for oxy-

polishing, which involves about energy penalty of around 5 MW. More importantly, the largest energy penalty is made up of the purification and compression of CO_2, which is about 30 MW. The optimization of operational energy penalty is crucial to improve the efficiency of large-scale CLC units, so energy integration and optimization will be a vital issue in the future.

4.3 Complex Process Flow

Moreover, CLC has a more complex process flow. Although high CLC performance is obtainable in lab-scale modules, the complicated reactor structure and operation process, as well as the lack of design theory and operation specification, all become the bottleneck issues faced by the scaling-up of the CLC technique [10]. Unlike traditional combustion technology, CLC is usually performed in interconnected fluidized beds, and additional facilities such as steam generation, char separation, and FR flue gas combustion are required to assist its operation.

In addition, the two reactors in CLC technology are coupled and affected by factors such as solids circulation and pressure balance, which makes the system not flexible enough. To meet the demand for lattice oxygen and heat transfer between the AR and FR, a high solid circulation rate and high solid inventory are needed to guarantee sufficient contact and reaction between the OC and fue, increasing the operating cost and making it more difficult to realize a stable operation, resulting in unstable fluidization, empty loop-seal (LS), and inverse gas flow of the LS [11]. Besides, more severe sintering and the agglomerate problem of the OC particles will occur due to high mechanical, chemical, and thermal stresses related to circulation in the process loop during life-time, which often bring some operation problems, such as de-fluidization, plugging the distributor and eventually, total collapse of the fluidized bed

Table. 1. Comparison of power generation with different carbon capture processes

Parameters	Without capture	Post-combustion capture	Oxy-fuel combustion	Coal staged gasification	CLC
Input MW	1397.2	1397.2	1397.2	1397.2	1397.2
Efficiency/%	42.3	33.9	38.8	41.7	42.2
E_{ad}/% (additional fuel consumption, without CO_2 purification and compression)	/	24.7	9.1	1.4	0.3
E_{ad}/% (with CO_2 purification and compression)	/	34.8	14.5	7.9	7.4
Efficiency penalty/%(with CO_2 compression)	/	10.9	5.4	3.1	2.9

5. POTENTIAL OPPORTUNITIES

5.1 Significant Cost Advantage

The energy penalty is the primary cost driver of CO_2 capture. Additional energy demand for the separation process results in significant decreases in efficiencies of pre-combustion capture, post-combustion capture, and oxy-fuel combustion technologies, ranging from 6-11%, 9-14%, and 7-11%, respectively [12]. This has resulted in a setback of nearly half a century in terms of current power generation efficiency, which is economically unacceptable.

CLC technology is more competitive than traditional CO_2 capture technologies in reducing energy penalties and costs [13]. This is because its ability of CO_2 inherent separation allows for high-concentration CO_2 enrichment at the source, which avoids any gas separation process. Consequently, it can theoretically achieve really low energy penalties mainly associated with CO_2 compression. As shown in Table 1, CLC technology has the lowest efficiency penalties (2.9%) and lowest additional fuel consumption (7.4%) in the case of CO_2 purification and compression and exhibits a similar power generation efficiency as the base combustion technology without carbon capture. As discussed in section 3.1, it also shows the lowest cost among other technologies precisely due to the high power efficiency. In the CO_2 capture development roadmap formulated by the U.S. Department of Energy, CLC technology demonstrates the highest carbon capture efficiency and cost benefits

5.2 Expected Leaping Development

Over the past 20 years, significant progress has been made in reactor design/construction/operation and OCs development in CLC technology, providing a firm base for the transition from laboratory-scale to industrial demonstration of CLC technology. For CLC reactors, there are primarily two stages of development based on reactor scale: the laboratory fundamental and the pilot demonstration plant research stage. To date, there are now over 40 CLC test units worldwide, ranging from small-scale laboratory scale (0.2 kW$_{th}$) to semi-commercial (4 MW$_{th}$), with a total operation time of more than 10000 h. The highest 4 MW$_{th}$ CLC unit has accumulated over 1000 hours of testing strongly confirms the realism of CLC for large scales [14].

As for OCs, researchers have conducted CLC performance tests on over 2000 existing OCs. CLC systems using synthetic OCs have accumulated over 8000 h of operational experience due to their high reactivity. It has been found that synergetic reactivity can be achieved via a mixture of different kinds of low-cost materials, addressing the issue of relatively low reactivity compared to synthetic OCs. Additionally, simple and effective particle mixings methods, such as cement bonding, spray drying, and extrusion-spheronization, have been developed for the large-scale preparation of low-cost materials, reducing the production cost of OCs.

The rapid development of CLC technology leads to a wealth of operational experience. As shown in Fig.3, currently, CLC technology stands at a technical readiness level of 6 [8], and it is expected to gradually move towards industrial demonstration in the next decade.
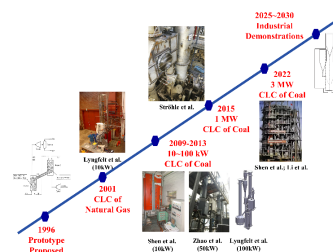

Fig. 3. Development of the CLC units

5.3 Value-added Chemical Production

As a highly versatile and effective process intensification strategy, the chemical looping (CL) concept refers to decoupling the overall reaction into multiple sub-reactions that occur in different spatial or temporal domains, with solid intermediates that can be regenerated to close the loop. With the extension of the CL concept, various chemical looping strategies beyond combustion (CLBC) have been studied over the past few decades, especially the production intensification of value-added chemicals [15].

Unlike the complete oxidation and the transformation of chemical energy to thermal energy in CLC technology, the CLBC process converts various fuels into desired chemicals via chemical looping partial oxidation (CLPO), thus upgrading the low chemical energy to high. Moreover, this process avoids inefficient separation steps through in-situ product separation and cascaded energy utilization, which have less exergy loss and high utilization efficiency of lattice oxygen. CLBC are often associated with the selective conversion of reactant molecules. When appropriately designed, the OC can be compatible with various oxidizing and reducing agents, so it is possible to selectively oxidize light alkanes, aromatic hydrocarbons, and biomass to target products.

From a global perspective, if suitable carriers that allow the transport of desired functional groups can be found, the principle of CL should fundamentally apply to

any catalytic reaction. The inherent advantage of reactant and product separation in CL technology may have a significant impact in the future on a broad range of chemical processes, yielding higher efficiency and "greener" chemistry.

6. REMAINING CHALLENGES

6.1 Balance the OC Performance and Cost

The development of OCs with high performance and low cost has always been a focus in CLC technology. The relationship between the reaction performance and cost of several representative OCs is roughly given in Fig. 4. It can be observed that highly reactive OCs are usually expensive. Therefore, achieving a good balance between the cost and reaction performance of OCs is crucial. This is typically achieved through the following approaches: (1) the large-scale production of low-cost and high-performance OCs through the synergistic effects between different cheap materials, and (2) enhancing the reactivity and stability of OCs through tailored design.
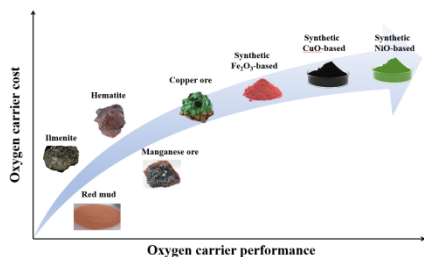

Fig. 4. Relation between the reaction performance and cost of OC materials.

Yang et al. [16] conducted a simple mechanical mixing of copper ore and iron ore, resulting in a composite OC that demonstrated synergistic effects in improving the reactivity of iron ore, as well as resistance to agglomeration of copper ore. Furthermore, Su et al. [17] and Dong et al. [18] achieved the batch preparation of composite OCs by spray drying and extrusion spheronization methods, which exhibited good stability during the redox test, successfully achieving the large-scale production of low-cost materials.

To enhance the anti-agglomeration performance of OCs, a core-shell $CuO@TiO_2-Al_2O_3$ hierarchical structure is synthesized as an OC using the self-assembly template combustion synthesis (SATCS) method, which enabled the high CuO-content OC could achieve high stability and combustion efficiency (>97.2%) over 20 cycles [19]. Focusing on preventing the phase transition of OCs, Liu et al. [20] prepared highly stable and reactive $CaMn_{0.5}Ti_{0.375}Fe_{0.125}O_{3-\delta}$ (CMTF_Lab) at the laboratory scale. Compared to conventional OCs, whose oxygen decoupling process is accompanied by phase transition, CMTF_Lab exhibited faster oxygen uncoupling and redox kinetics. Then they [21] further upscaled the production of CMTF_Lab to the industrial level (CMTF_Ind) through spray drying. The experiments demonstrated that CMTF_Ind exhibited a total oxygen transfer capacity of 5.71 *wt.%* and excellent stable redox performance.

6.2 Manage Heat and Mass Transfer and Facilitate the Rate-limiting Step

Effective heat and mass transfer management is crucial to design and to operate a CLC unit. With this regard, the potential solutions can be proposed from two aspects, i.e. from the OC and reactor standpoints. For scaling-up dual CFB reactors, the solid circulation rate calculated based on the heat balance is usually greater than the value based on the mass balance. To reduce the solid circulation rate, an efficient method is to use a kind of OC that makes FR exhibit overall thermal neutrality or slightly exothermic [22]. Additionally, the circulation of OC is the key link in regulating the operation of the CLC reactor, which maintains the mass and heat transfer between the AR and FR. However, the dual-bed reactor configuration increases the difficulty of controlling the solid circulation rate, and it is easy to encounter the problem of mismatched circulation rates between the AR and FR. To address this issue, a bidirectional loop seal can be used to establish the solid circulation between the FR and AR and the internal circulation of the FR, which can attain more flexible control of solid circulation.

Furthermore, as mentioned above, the slow gasification of coal char is the rate-limiting step in the *i*G-CLC process of coal. Regulating and promoting the coal char conversion process is a crucial issue in iG-CLC technology. With this regard, the potential solutions can be proposed from two aspects. First, from the OC standpoint, using the OC capable of releasing gaseous oxygen (such as copper ore, manganese ore, and Cu-decorated iron ore) can significantly increase the coal char conversion rate [23]. Additionally, alkali/alkaline earth metals have been proven to increase the coal char gasification rate in the *i*G-CLC process [24,25]. Besides, from the reactor standpoint, prolonging the residence time of coal char in the FR can also promote its gasification, which can be achieved by optimizing the reactor configuration, including changing FR structure (such as multi-stage/multi-chamber FR, internal solid circulation in the FR) [26] and using supplementary devices (such as carbon stripper, secondary FR) [27].

6.3 Matured Design Theory and Operation Methods of Reactor

The design theory and operation methods of the CLC reactor are the basis for CLC technology implementation. However, because the current research lacks experimental evidence of pilot-scale testing in the thermal mode, there is a lack of mature design theory and operational specification. In this case, researchers have conducted numerical simulation methods, such as CFD simulation and macroscopic modeling, into the design and operation of reactors to help scale up the CLC unit to the commercial demonstration.

The application of numerical simulation methods depends on advanced and reliable models, among which mesoscale models serve as a bridge between macroscopic fluidization models and microscopic reaction mechanism models. Su et al. proposed a new inter-phase drag model in which heterogeneous characteristics of particle clusters in fast fluidized beds from mesoscale particle aggregation and interfacial structure [28] and established a particle surface reaction kinetic model via particle-scale simulation in CLOU [29]. These mesoscale models can be coupled with CFD simulations. Furthermore, numerical simulation can provide valuable references for engineering design and offers assistance in operational parameter optimization. Chen et al. optimized the design of a 50 kW$_{th}$ dual CFB reactor for CLC of coal using the Computational Particle Fluid Dynamics (CPFD) method. As optimizations, well-mixed gas-solid contact was achieved with two oppositely-collocated coal feeding points [30], and higher carbon capture efficiency and combustion efficiency can be obtained with a new four-chamber device coupled in a LS [31] (Fig.5).
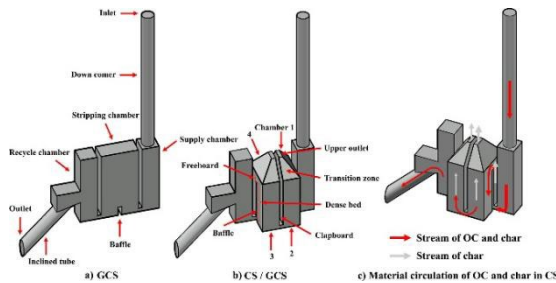

Fig. 5. Structure of the (a) original and (b) optimized CS [31]

7. SUMMARY AND PERSPECTIVE

CLC is an innovative combustion and carbon capture approach used in the utilization of coal and other high-carbon fuels, which enables cascaded energy utilization and *in situ* pollution control. However, it is currently limited by challenges such as mismatched reaction rates, high cost and short lifetime of OCs, and difficulty in regulating heat and mass transfer and chemical reactions in the reactor, which restrict its achievement of high combustion efficiency, CO_2 capture efficiency, and scale-up.

In terms of OCs, developing cost-effective while well-performing OCs is a key focus of future research. Machine learning methods can facilitate efficient and low-cost trade-offs between performance and cost by using automated or parallelized techniques for OC property prediction and high-throughput screening. Additionally, establishing a unified performance test standard for OCs is essential to assess their potential applications in an actual CLC process based on test results. Regarding reactors, the critical challenges are achieving high combustion and CO_2 capture efficiency simultaneously and realizing continuous autothermal operation. Numerical simulations based on macroscopic modeling or commercial simulation software (such as CFD) can provide helpful theoretical guidance for reactor design and operation optimization. Furthermore, it is very urgent to validate the feasibility of MW scale CLC pilot units for long-term continuous and stable operation in the thermal state to provide practical experience for the reactor design.

China's "National Science and Technology Innovation 2030 Major Program" has identified CLC technology as one of the important research directions for CO_2 capture technology, which signifies an unprecedented opportunity for promising CLC technology in China. Therefore, despite certain obstacles to the development of CLC technology, we believe that with the joint efforts of research institutions, government, and investors, it is reasonable to anticipate the successful implementation of CLC on an industrial scale very soon.

DECLARATION OF INTEREST STATEMENT

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. All authors read and approved the final manuscript.

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