The Efficient Fuel Combustion Using Chemical Looping

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Abstract. This paper discusses Chemical Looping Combustion (CLC). That is the system for energy generation and pollutant control which can capture carbon dioxide (CO_2). CLC uses the carrier of solid oxygen that can transfer the oxygen from air to fuel during combustion, eliminating the need for an air separation unit (ASU), and allowing to production of an approximately pure stream of carbon dioxide for the purpose of capturing and storing. The paper also discusses the history of CLC, the types of fuels that can be used in CLC, and the challenges and difficulties this technology is facing. The second part of the paper centers on the topic of different types of solid oxygen carriers, including copper oxides, manganese oxides, and iron-based oxygen carriers. The paper also discusses utilizing solid fuels in CLC systems, particularly coal. All the essential components required for expansion are presented and accounted for. Various inexpensive oxygen transport substances have undergone extended trials and have exhibited satisfactory levels of effectiveness and durability. Overall, CLC is a promising technology for reducing the environmental impact of energy production and industrial processes.

Keywords: Chemical Looping Combustion; large models; Oxygen Carriers; Biomass.

1. Introduction

Chemical Looping Combustion, also called CLC, is a system that can capture carbon dioxide (CO_2). It is an emerging technology for power generation and pollutant control. It can be used in a variety of industries, such as power generation, steel, and cement. CLC shows potential as a substitute for traditional combustion techniques because it allows for more efficient and cleaner combustion of fossil fuels. A solid oxygen carrier is utilized in CLC that transfers oxygen from the air to fuel during combustion, thereby eliminating the need for the air separation unit and allowing for the production of a nearly pure stream of carbon dioxide for capturing and storing, which is called Carbon Capture and storage (CCS).

The earliest concept of Lewis and Gilliland proposed CLC in 1954. Then, Richter and Knoche in 1954 and Ishida in 1983 did more research about CLC. In 1987, Ishida has drawn more research attention to the Clean Low-temperature Combustion (CLC) process deserves greater research focus as a potentially effective method of producing clean energy. CLC consists of two reaction steps in a sequential cycle, using the use of carriers of solid oxygen in interconnected fluidized bed reactors. That is accomplished by employing metal oxides [1].

Just like one of the most basic CLC models shown in Figure 1, there are two reactors in this system, and one is a fuel reactor. That is where people put the fuel in that container, and then the fuel will burn in this container. Then the fuels will turn to steam in another container called an air reactor. In chemical cycle combustion, gaseous fuels are burned by utilizing an oxygen carrier, usually a metal oxide. Oxygen is transferred from the combustion air to the fuel without direct interaction between the fuel and combustion air. This leads to the automatic separation of the greenhouse gas CO_2. As a result, the combustion products, such as H_2O and CO_2 are segregated from the remainder of the flue gas, and following condensation, almost entirely pure CO_2 is produced with no energy waste on separation. This is the typical operational mechanism of the CLC system.

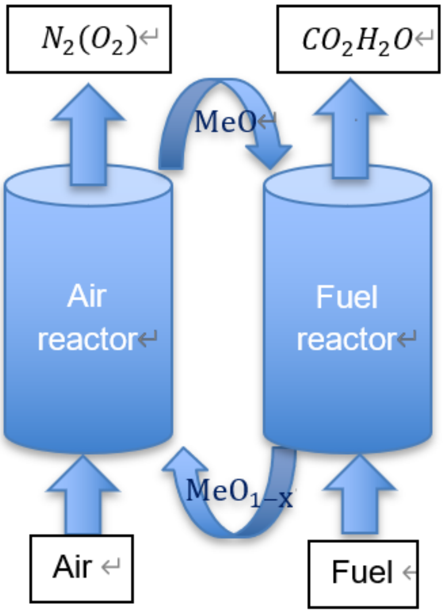

Fig 1. Metal oxide, typically MexOy, is used as a circulating agent to enable the combustion of fuel in a more efficient and cleaner manner [2].

There are three types of fuels that people can use, which are solid fuels, liquid fuels, and gas fuels. The most widely known fuel is solid fuel. When the solid fuels go inside fuel reactor, they will release the volatiles and the char. The oxygen carrier will directly react with volatiles. The char will form the steam gasification to syngas such as H_2 and CO. ideally, individuals would aspire for the fuels to undergo complete combustion within the fuel reactor, subsequently resulting in complete conversion to carbon dioxide and water, akin to the illustration depicted in Figure 2. However, in the case of solid combustion, it is usually not possible to achieve complete combustion, resulting in the fuel not being able to reach the full amount of gas to be spent. The addition of oxygen to the downstream post-oxidation chamber following the fuel reactor, also known as oxypolishing, can serve as a solution for this issue [2].

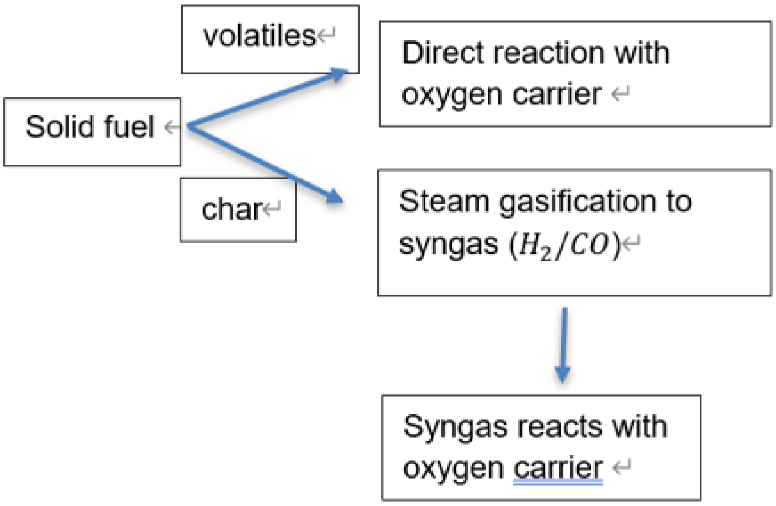

Fig 2. Chemical reactions involving solid fuel in Chemical Looping Combustion (CLC) [2].

Under ideal conditions, that will be the dissipation Comprised of combustible gases such as H2, CO, and CHx when gas leaves fuel reactor. That shows the incomplete gas conversion. Also, there will be the depletion of char through air reactor and its loss along with gas that exits fuel reactor [2]. Similar to illustration presented in Figure 3. That means the incomplete CO_2 capture.

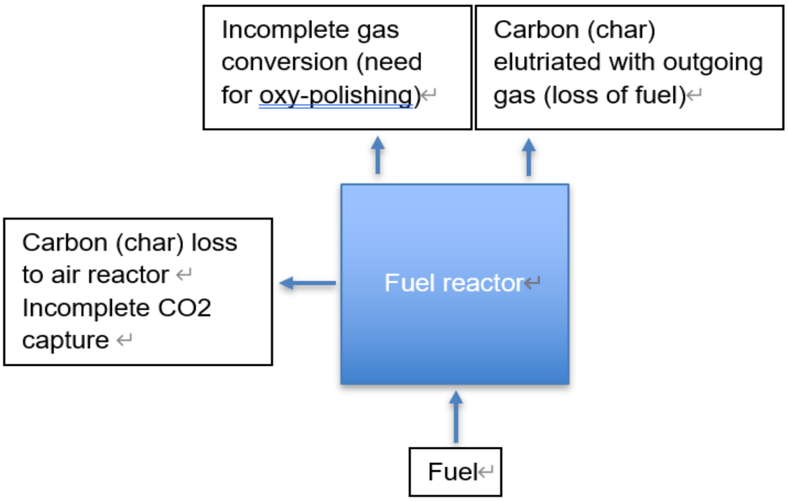

Fig 3. Insufficient functioning of the fuel reactor [2].

Now, this technology still faces many difficulties and challenges. For example, this technology is very prone to pollution. These pollutants can easily produce greenhouse gases that lead to global warming. Individuals endeavor to discover methods to alleviate pollution; however, the task is not as straightforward as initially perceived. However, CLC is a brand-new technology, so there is investment risk. People need to take the CLC technology to scale. Evolving from small size to large size is not a simple matter. There will be a lot of uncertainty with the large size. The design of the device is also very different. Data from small-scale experiments cannot be fully applied to large-scale experiments, and there are many uncertainties in large experiments. One would also consider the cost, as the cost of maintenance and materials required for this technology is quite expensive. One needs to find ways to reduce the overhead as much as possible [2].

Overall, CLC is a promising technology for reducing the environmental impact of energy production and industrial processes. Its applications anticipated to increase in the near future as the need for more sustainable and efficient energy solutions becomes increasingly urgent.

2. Oxygen-carrying materials

Oxygen-carrying materials are a crucial component of CLC technology. In CLC, instead of directly burning fuel with air, the combustion process is facilitated by utilizing the oxygen carrier that facilitates the transfer of oxygen to the fuel.

Table 1 shows results for 14 different calcium manganate-based materials. Those materials continue to be tested in the CLC systems. The creation of spherical particles, ranging from 100-200 µm, is achieved through spray drying and subsequent sintering. For a material to qualify for testing in a 300 W device, it must exhibit notable reactivity in the batch experiments involving some of grams of oxygen carriers and possess high mechanical wear resistance. In order to be considered appropriate for use in 10 kW unit, the material must show minimal levels of wear and deterioration, in addition to a substantial fuel conversion rate, which is established during the continuous testing of 300 W unit [3].

Table 1. Summary of oxygen carrier materials examined and duration of fuel exposure [3].

	Oxygen-carrier material notation	Sintering temp. (°C)	CLC tests conducted (h)	
			300 W	10 kW
[A]	C14-T(reference)	1300	16	55+41
[B]	C28-E1A	1350	35	-
	C28-C2A	1350	38	-
[C]	C28-E3	1280	7	-
	C28-E5	1310	3	-

2.1. Solid Oxygen Carriers

Selecting the appropriate oxygen carriers is a crucial aspect of the CLC system. Oxygen carriers that are made of transition metal oxides, including copper, iron, manganese, and nickel, are considered highly promising because they can be easily reduced and reoxidized.

To optimize the effectiveness of oxygen carriers in CLC, several essential properties must be considered. These include the ability to quickly and efficiently transfer oxygen to the fuel, maintain stability throughout the CLC cycle, resist the mechanical stresses related to cycling, avoid clumping together, have a minimal environmental impact, and be affordable. Additionally, other factors, such as thermochemical stresses and mechanical forces, impact the lifespan of oxygen-carrying particles during multiple cycles of reduction and reoxidation. Therefore, the power of the oxygen-loaded particles is critical due to the mechanical strains they encounter during fluidization [4].

2.1.1. Copper oxides

One good example of Solid Oxygen Carriers is Copper oxides (CuO). Copper oxide is a promising the high reactivity with methane. This property renders it a fitting oxygen carrier for CLC with good regeneration rate and relatively low cost. Moreover, the reduction and reoxidation of CuO are exothermic reactions that obviate the necessity for supplemental energy. However, the use of CuO in CLC is limited by its low decomposition temperature and tendency to agglomerate. Researchers have investigated the use of supported CuO, which shows improved stability compared to bulk CuO. However, the chemical interaction oxygen carrier with the supporting material can limit the amount of reducible phase in the supported oxygen carrier, resulting in the formation of non-reactive phases. Instances of this phenomenon are evident in the creation of $CuTiO_4$ and $CuAl_2O_4$ as TiO_2 and Al_2O_3 are employed as a backing for CuO oxygen carriers [4].

2.1.2. Manganese oxides

Another example of Solid Oxygen Carriers is oxides of manganese. Such as, MnO2 and Mn3O4 are renowned for their capacity to engage in reactions with methane. Nonetheless, their decomposition restricts at low temperatures their applicability in CLC. Recent studies have explored the use of mixed manganese oxides with other transition metal oxides, such as CuO, which have demonstrated promising activity for syngas fuel oxidation. Additionally, manganese ore has recently garnered interest. Ilmenite has been proposed as an additive to prevent the decomposition of the oxygen carrier during its application. In research conducted by Frick, the performance of 18 various mixed ratios of Cu-Fe and Ni-Mn oxygen carriers was evaluated, with the Ni-Mn mixture comprising 34% NiO exhibiting the highest efficacy. This mixture demonstrated complete conversion of CH4 to CO2 while maintaining good mechanical integrity [4].

2.1.3. Iron-based oxygen carriers

Fe-based materials offer several benefits, such as affordability, non-toxicity, resistance to sulfur, and adequate heat resistance throughout the process. While Fe_2O_3 has the potential to be converted to Fe when combustible gases, for example the presence of CH_4, CO, and H_2, within the integrated fluidized bed CLC system, thermodynamic or kinetic constraints limit the process of reducing Fe_2O_3 to Fe_3O_4. If Fe_3O_4 is further reduced to Fe or FeO, it may result in a reduction of the purity of CO_2 produced in the FR due to the increased equilibrium concentrations of CO and H2 in the flue gas. Furthermore, the profound decrease in the oxygen transport agent based on iron could lead to particle sintering and agglomeration, highlighting the need to restrict reduction beyond Fe_3O_4 in the CLC process [5].

The decrease of Fe_2O_3 using the products obtained from the thermal decomposition of coal and the conversion of the remaining char into gas was examined by pretreating commercial Fe2O3 powders at 1173 K for 5 hours, and then utilizing them within a reactor with a stationary bed for the CLC of low-rank coal, oxygen serves as the carrier. In contrast to commonly used experimental setups. In this study, quartz wool was used to separate coal samples and Fe_2O_3 particles, which were mixed together in the form of solid fuel and particles that serve as oxygen carriers. The products of either the thermal decomposition of coal or the conversion of char into gas introduced onto Fe_2O_3 for reaction using a carrier gas. Similarly, Zhao and colleagues proposed the reactor with fluidization taking place in two stages for differentiating the destiny of mercury and sulfur during the sub steps of thermal decomposition of coal and conversion of char into gas in coal-based integrated gasification CLC. This two stage reactor layout separates coal and oxygen carrier samples into the reactors of the initial and subsequent stages, respectively, facilitating clear differentiation of emission features of pollutants during the stages of coal pyrolysis and char gasification [5].

2.2. Solid Fuel

The cheaper fuel is solid fuel because that is much easier to get like coal and waste. People use different sizes of coal putting in different power of CLC systems. First, using two quite different sizes of coal were put into a 1 MW pilot. One is pulverized coal. The result is shown in figure 4. It shows a kind of low present of performance on carbon capture. That may be because the coal cannot burn completely in the fuel reactor. The expense incurred results in a significant loss to the air reactor. The result shows that carbon capture is only at 80 percent. A considerable amount of carbon is lost from the fuel reactor through elutriation. The result shows that the fuel conversion is only 50 presents. Another size of coal is coal particles with a diameter of up to 8 mm. When it is in the CLC systems, a significant amount of char is lost to the air reactor, which shows in the data that the carbon capture is even lower than 50 percent. That may be because the larger size of coal has a shorter time that stays in the fuel reactor. They cannot burn completely over there, which means the fuel reactor is really hard to transform the bigger coal particles. When the pulverized coal in power is at 100 kW unit, the carbon losing through elutriation is still high, but that is better than the 1 MW unit. The percent of the performance of fuel conversion is about 35 percent higher than 1 MW. The reason for this result is the low operating temperature of the test unit in the 1 MW unit, which resulted in the incomplete combustion of the coal in the fuel reactor to attain full conversion. In addition, there is a low carbon loss to the air reactor. In the 100-kW unit, which means it has a higher percentage of the performance of capturing carbon. Furthermore, the gas conversion is marginally enhanced in the 100-kW unit. The results for the 100-kW unit after the addition of manganese ore show similar results, except for a slight increase in gas conversion. The test of 90-300 μm of the coal which is the medium size of the coal in 100kW unit showed a higher percentage of the performance of fuel conversion efficiency, but it does not help with the elevation of the loss to the air reactor. The addition of manganese ore to the process explains good gas conversion. In the 50kW experiment, one also used medium-sized coal. Running at 13.5 kW also shows lower elution losses. In addition, compared to the 100-kW unit, the 50 kW unit displayed satisfactory gas conversion, but the losses in carbon conversion to the air reactor were still slightly high [2].

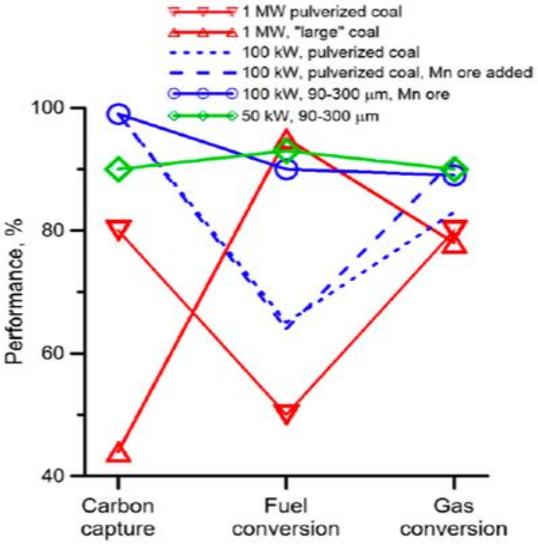

Fig 4. Evaluation of the operational efficiency across three experimental coal-based pilot studies [2]

In summary, it is clearly implied that using medium-sized fuels is a very good choice because if fuel pellets are excessively big, they will not possess enough time to fully burn in combustion reactor and thus fully convert, and then the incompletely burned fuel will follow the circulating flow directly into air reactor. If the particles are excessively tiny, they may follow the air stream directly and elute quickly, and then be lost.

2.3. The Entire Process of CLC

Preliminary tests were conducted to evaluate the impact of immediate contact between coal and oxygen carrier (OC) on coal carbon conversion. In one experiment, the coal and OC were combined and inserted into the first stage fluidized bed reactor. In contrast, coal was inserted into the reactor that had a fluidized bed system arranged in first stages, while the OC was filled into the second stage fluidized bed reactor in the other experiment, preventing direct contact between the two. The instantaneous carbon conversion rates were determined to be$7.491 \times 10^{-3}$and $7.212 \times 10^{-3} \mathrm{s}^{-1}$ for the first and second tests, respectively, indicating no significant difference in the carbon conversion rates with and without OC and coal in direct contact. The final reaction temperature, heating rate, and coal type were discovered to be the primary factors affecting the fast pyrolysis of coal, rather than the contact with OC. Nonetheless, test No. 2 demonstrated improved gas-solid mixing conditions, resulting in a higher tar conversion rate [6].

Table 2. Evaluation indexes of CLC [6].

No.	$rx_c = 0.4(s^{-1})$	Y_{CO2}	Y_{CO}	Y_{CH4}	η_c	Carbon balance
1(mixed)	7.491×10^{-3}	98.080 ± 0.010	1.382 ± 0.005	0.542 ± 0.006	98.798 ± 0.007	1.051 ± 0.007
2(separated)	7.212×10^{-3}	99.698 ± 0.005	0.161 ± 0.003	0.143 ± 0.002	99.801 ± 0.004	1.103 ± 0.003

In test No. 1, where a mixture was used, a 98.080% CO_2 yield, which was superior to the yield achieved with hematite like OC. Nevertheless, during the No. 2 test, where the conditions were altered, there was a slight increase of 1.618% in CO_2 yield and 1.03% in combustion efficiency. The carbon balances for both condition No. 1, in which the components are mixed, and condition No. 2, in which the components are separated tests were satisfactory, with average values of 1.051 and 1.103, respectively [6].

3. Chemical Looping Combustion Changes from Small to Large Models

It is very difficult to change from CLC in the small model to CLC in the large model because the data in CLC in the small model are completely inapplicable in the large model. “The fuel reactors with 100 kW and 1 MW capacity have heights of 5 m and 9 m, respectively. A complete reactor on a larger scale could have a height of 50 m, which would allow for extended residence time to facilitate char conversion [2].

The 1000 MWth design differs from the 100 MWth design as it does not have a carbon stripper and has a different method of sending the material that circulates from the fuel reactor back to the air reactor. However, both designs share similar principles. Despite being smaller, the 100 MWth design has eight cyclones and lower riser heights, and lower velocities compared to the larger unit. This results in a greater cross-sectional area per unit of power. In 100 MWth design, the carbon stripper adds to the fuel reactor's cross-sectional area. There are various designs for small CLC pilots, but not all are applicable for full-scale units, and it is advantageous to draw from the experiences of CFB technology [2]. Therefore, it is still a question of how to bring CLC to scale.

4. The Packed Bed Process for the Chemical Looping

The CL_PB process, which employs packed bed reactors, is used for reforming natural gas and comprises three conversion units that enable gas-solid and catalytic reactions. Unlike the linked fluidized bed, this system is operated dynamically. The gas solid reactor with a packed bed enables faster movement of the reaction front towards the reactor's end, which may cause heat to accumulate within the reactor. This heat can be extracted by generating high-temperature gas to be used for generating power or by implementing a reaction that absorbs heat, such as steam reforming, when an exothermic reaction, such as oxidation, is present [7].

5. Biomass in CLC

CLC has potential benefits in the case of biomass. Among them, a significant advantage is a potential for low or zero emissions of NOx. It is the least environmentally damaging fuel that can be found, as it can be more easily burned in a fuel reactor, thus producing minimal particulate emissions into the air.

Another potential advantage of using CLC for biomass is related to the challenges posed by corrosive components of alkali ashes that are present in biomass fuels. In case, the alkali can be prevented from entering air reactor, that could lead to reduced maintenance costs and improved efficiency. It could also enable the use of biomass fuels that are typically avoided in boilers due to their high potassium content, such as grasses [8].

The presence of certain ash components can cause particles in the bed to stick together and form clumps, which can lead to the bed material being unable to be fluidized. This is known as defluidization. To investigate this issue, Zevenhoven and colleagues conducted a study using salts to simulate biomass ash in a laboratory reactor. The results of their study were deduced that KCl would undergo vaporization within the fuel reactor, thus preventing presence of alkali chloride in air reactor [5]. Given large number of fluidized bed boilers currently in operation that utilize biomass, bio-CLC is a highly suitable option for early implementation of the technology. Following the up-scaling approach described earlier, which involves utilizing an existing CFB, bio-CLC has a strong potential for successful implementation at scale [8].

6. Alkali Release during Fuel Conversion

In the gasification phase of CLC, residual coke undergoes steam gasification, and the resulting gases and volatiles react with oxygen carriers. The liberation of K during this process in CLC has not been investigated earlier. However, gasification conditions in CLC system are comparable to those in the traditional steam gasification, which entail high temperature, an oxygen-free atmosphere, and surplus steam. In traditional steam gasification, the primary mechanism for KCl release into the gas phase is evaporation, although some KCl (g) is transformed into KOH (g) and HCl(g) due to steam reforming. In the gasification stage of CLC, the expected net K release from char, like KCl (g) or through steam reforming to KOH (g), through KCl evaporation is projected to resemble traditional steam gasification and combustion. Therefore, it is insufficient to account for the observed high release of K into the flue gas [9].

The increased release of alkali observed in CLC can be explained by Novakovich et al.'s experiments, which demonstrated that water vapor's presence considerably accelerated the thermal breakdown at high temperatures of K_2CO_3. This phenomenon was ascribed to the reaction of K_2CO_3 with steam resulting in the production of KOH (g). This phenomenon was ascribed to the reaction between K_2CO_3 and steam, producing KOH (g). Likewise, investigations of ilmenite reaction with the K_2CO_3 and K_2SO_4 demonstrated that the vapor's presence in the reaction environment boosted K's emission to the gas phase by forming KOH (g). The molar ratios of K to Cl in the fuels used in this study were 6.93 for wood pellets, 14.01 for charcoal fuels, and 1.88 for pellets made from straw. Assuming that for both highly volatile fuels, like pellets made of wood and straw, a significant fraction of Cl is released during devolatilization, it suggests that the remaining char fraction's K to Cl ratio is higher, and a considerable fraction of the fuel's K may exist as SO_4 salts, such as K_2SO_4 and K_2CO_3. Hence, the steam used as the fluidized substance utilized in the CLC procedure could accelerate the disintegration of K_2SO_4 and K_2CO_3 salts, leading to a noteworthy discharge of K like KOH gas [9].

7. The Cost of Energy Associated with Separating and Compressing Gas.

The cost of capturing CO_2 involves two main processes: separating CO_2 from other gases at ambient pressure and compressing the CO_2 to a pressure suitable for transport and storage. The theoretical energy required to compress CO_2 from 1 to 100 bar can be calculated using the gas constant, temperature, and initial and final pressures. Assuming ideal gas behavior and isothermal conditions, the theoretical work of compression is 0.255 MJ/kg of CO_2. However, non-ideal behavior at higher pressures, especially at lower temperatures, the compression energy required is actually higher than this value.

Energy necessary for the gas separation is contingent on the CO_2 concentration in the gas mixture. When dealing with the CO_2 concentrations commonly found in coal and biomass fuel combustion, the energy required for separation is less than half of the energy needed for compression. Nevertheless, when dealing with direct air capture, the energy necessary for separation exceeds three times the amount required for combustion sources.

The primary energy cost associated with the MEA process for gas separation is heat required to regenerate the absorbent, which is considerably greater than the energy needed for compression. However, the regeneration energy for the KS1 solvent utilized in the Petra Nova facility is less than that of MEA. In the power plants, the steam at a reduced pressure produced can be harnessed to produce the thermal energy required for solvent restoration, thereby reducing the electricity cost.

In the case of oxyfuel, the energy needed to separate gases, assuming high purity, is approximately 0.9 MJ/kg of O_2. For coal and biomass, this corresponds to 0.65-0.76 MJ/kg of CO_2. When coupled with compression, this results in an energy penalty of 0.98-1.1 MJ/kg of CO_2, which accounts for approximately 10% of the calorific value of coal or biomass containing moisture content of 20% [10].

8. Reduction of the Expenses while Obtaining CO2

In general, low-cost solid fuels are commonly paired with oxygen carriers due to potential losses during fuel ash separation. However, these carriers are not expected to achieve full gas conversion. Although there is an assumption that gas generated in fuel reactor can be entirely oxidized by oxygen, leaving some residual oxygen, it may be necessary to eliminate excess oxygen as well as sulfur oxides and nitrogen oxides. Concentrating sulfur oxides and nitrogen can facilitate their removal and reduce costs.

Recycling the remaining combustibles back to the fuel reactor after separating CO_2 from the combustible gas is a potential solution. The industry commonly employs the Benfield process to separate CO_2 present in the process gases, similar to the gases produced in the fuel reactor depicted in Figure 5 serves as the location for the following process. If fuels containing sulfur are used, the Benfield process can trap a gas blend containing CO_2, SO_2, and H_2S, necessitating the removal of the second and third before or after the process. If sulfur-containing fuels are used, the Benfield process can trap a blend of gases containing SO_2, CO_2, and H_2S. As a result, it would be necessary to remove the latter two either before or after the process. The Benfield process has several advantages. Firstly, it does not require oxygen production or extraction of the gas. Secondly, reusing gas can be an alternative to steam for fluidizing the fuel reactor, thereby reducing the energy cost related to steam injection [2].

Compared to oxygen addition to meet gas demand, the Benfield process is less dependent on the performance of CLC, making it advantageous, especially for high oxygen demands. Additionally, the energy needed for compression in the Benfield process gradually increases with the oxygen demand.

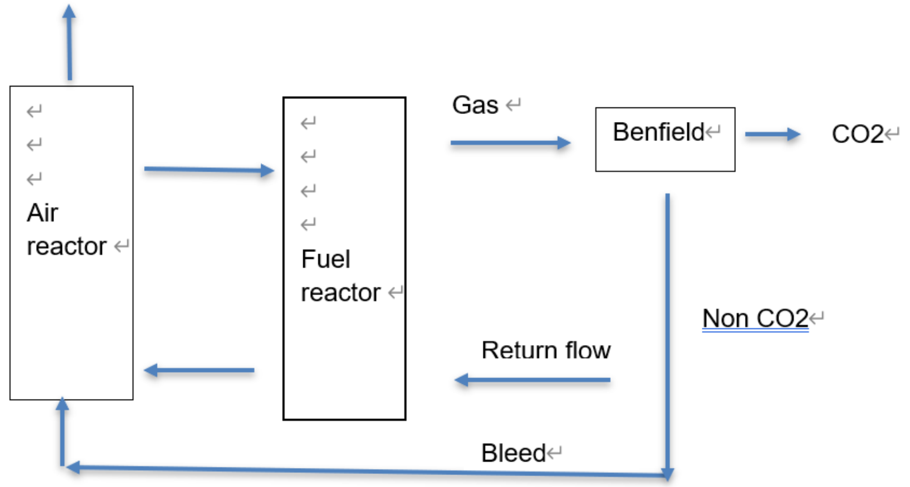

Fig 5. Evaluation of the operational efficiency across three experimental coal-based pilot studies [2].

9. Conclusion

CLC is a promising technology that enables efficient and clean power generation while also capturing carbon dioxide (CO_2). The process involves using solid oxygen carriers to facilitate the transport of oxygen from the air to the fuel while burning, resulting in a nearly pure stream of CO_2 that can be captured and stored. CLC operates using two fluidized bed reactors connected together, with metal oxides used like oxygen carriers. The system can handle solid, liquid, and gaseous fuels. Although the benefits of CLC are significant, challenges such as investment risk, pollution, and high maintenance costs still need to be addressed. Choosing the right oxygen carriers is critical to the success of CLC. This paper provides an overview of CLC's history, fuel types, challenges, and solid oxygen carrier options, for example, manganese oxides, copper oxides, as well as oxygen carriers that are based on iron. The utilization of coal and other solid fuels used in CLC systems is also addressed. With a range of affordable oxygen transport materials already tested and proven effective, CLC holds enormous promise for reducing the environmental impact of energy production and industrial processes.

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