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CHEMICAL LOOPING COMBUSTION AND CO_2 CAPTURE

TECHNICAL NOTE 26

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Summary

Chemical looping combustion (CLC) is an emerging technology that enables CO_2 capture without the high efficiency loss of current carbon captures technologies, in which metal-based oxygen carriers undergo repeated reduction/oxidation cycles to allow combustion without the fuel coming in contact with air.

The estimated efficiency of CLC using a gas turbine inlet temperature of 1200 °C has been estimated to be between 52 and 53 % (LHV) which includes the penalty for CO_2 compression to supercritical state. Co-generation of electrical power and hydrogen is also possible with CLC.

Research has been undertaken primarily in Europe, with efforts in the US and Asia. Experimental systems have been developed for gas fuels, and coal-based CLC is now being developed. The process has been tested in a gas fired 10kw prototype with semi-industrial scale demonstration (> 1 MW_{th}) of CLC being the next step in demonstration. Research has concentrated on system flowsheet assessments for efficiency optimisation, development of robust oxygen carriers, and prototype combustor operation to control operation with carrier transfer and leakage between reactors.

CLC can be seen as part of a family of technologies which includes membranes which, at present, are progressing through development based on gas fuels, with expectations to application with coal. CLC is more tolerant of the impurities associated with coal combustion than membrane systems, and could be therefore favoured. In addition, the technology uses a fluidised bed technology which is familiar to technology providers, even though this technology is not common in Australia.

Introduction

Combustion of fossil fuels in power generation is a major source of greenhouse gas emissions (1-7). Considerable effort is now being made towards developing more environmentally acceptable fuel use, partly through efficiency improvements and through the development of carbon dioxide sequestration technology. In combustion systems the interest lies in developing efficient and economic means for CO_2 capture. Three general approaches are available (4-7): Post-combustion capture, pre-combustion capture and oxyfuel technologies. Many comparative assessments of the capture techniques have been made, indicating efficiency is reduced by 10 – 25% and power costs increased by 25 – 75%, with the cost of carbon capture (not including CO_2 transport and disposal) estimated at US$ 20 – 50/ton CO_2. The need for air separation units (ASUs) for oxygen production in the technologies is a major contributor to cost increase and efficiency loss associated with carbon capture.

An alternative technology avoiding oxygen production is chemical-looping combustion (CLC) in which metal-based oxygen carriers undergo repeated reduction/oxidation cycles to allow combustion to take place without the fuel coming in contact with air.

In the oxidation reactor, the carrier reacts with oxygen in air as follows:

$$\mathrm{Me}_y\mathrm{O}_{x-1} + 0.5\mathrm{O}_2 \rightarrow \mathrm{Me}_y\mathrm{O}_x \qquad \text{- (1)}$$

In the reduction reactor, the carrier reacts with the fuel as follows:

$$(2n + m)\mathrm{Me}_y\mathrm{O}_x + \mathrm{C}_n\mathrm{H}_{2m} \rightarrow (2n + m)\mathrm{Me}_y\mathrm{O}_{x-1} + m\mathrm{H}_2\mathrm{O} + n\mathrm{CO}_2 \qquad \text{- (2)}$$

Reaction (1) is highly exothermic; reaction (2) can be slightly exothermic to slightly endothermic, depending on the carrier and the reaction conditions. The net heat liberated from both reactions is equivalent to that of normal combustion. The key advantage is that oxygen is separated from air without an energy-intensive ASU. The CLC process works in a cyclic manner illustrated on Fig 1 with continuous recirculation of oxygen carriers between an air and a fuel reactor, where oxidation (i.e. regeneration) and reduction (i.e. oxidation of fuel) of the metal oxide take place.

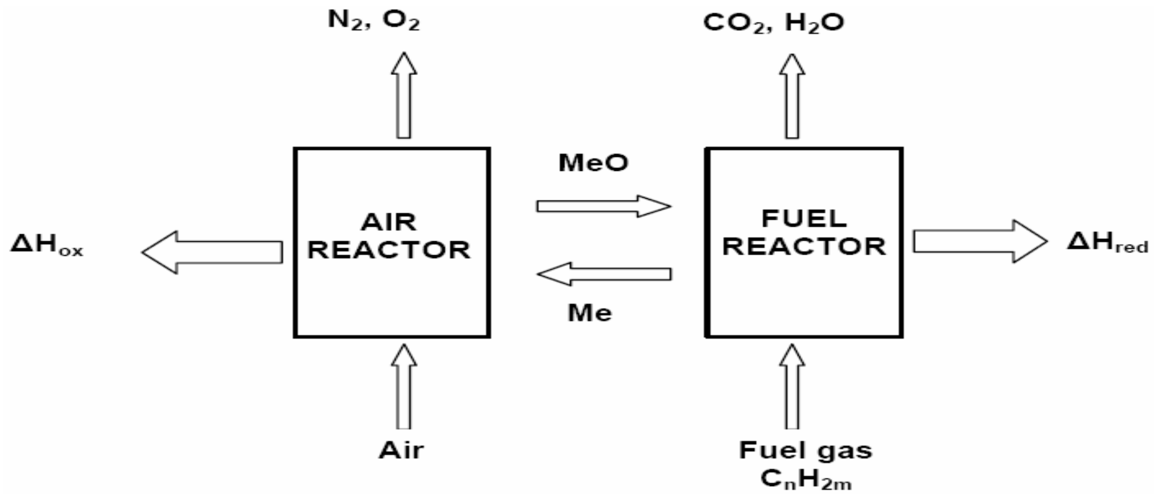

Figure 1: Schematic of the CLC process, with the oxygen carrier, Me, shown involved in the transfer between the air and fuel reactors

There are several additional issues. During reduction of the carrier, undesirable side reactions may also occur, leading to incomplete combustion. For example, formation of char on the carrier particles has been observed for many systems; this carbon must be burned off as CO_2 during the oxidation phase and released to the atmosphere.

Technology status and development prospects

Most research in CLC has been undertaken on the development of suitable oxygen carriers. The technology has been developed through concepts including assessments of costs and efficiencies, a 10kW prototype has been tested (8), and the next scale in development is considered to be at about 1 MW_{th} scale.

Most effort has been through EU projects. Chemical looping is one of the technologies considered in the EU ENCAP project (8) running from 2004-09, whose objective is to develop new pre-combustion CO_2 capture technologies and processes for power generation. It aims at technologies which meet a target of at least a 90% CO_2 capture rate and a reduction in the cost of capture of 50% compared to present. In the project CLC will be developed for gaseous and solid fuels and operated at atmospheric and pressurized conditions. *R&D* is directed towards materials and conceptual CLC-processes. The project aims to develop a 400 MWe SC CFB boiler for solid fuels CLC application.

Recent work incorporating gas turbines with CLC (9) predicts a low cycle efficiency penalty of 2%, mainly associated with CO_2 compression, and a CO_2 mitigation cost lower than 10 €/ton CO_2.

All experiments to date have been conducted with gaseous fuel, primarily methane. Carrier stability and cost has been assessed. Experiments at Chalmers University (8) for a CLC operated for 105 hours using a Ni-based oxygen carrier achieved 99.5% CH4 conversion with almost total selectivity to CO_2 and H_2O. There was no detectable gas leakage between reactors and the oxygen carrier suffered no loss in reactivity or particle strength. Extrapolation of data on fines lost during the test suggested that the lifetime of the carrier particles could exceed 40,000 hr. Based on a lifetime of 4000 hr the carrier cost was estimated at less than $1 per tonne of CO_2 captured.

There is considerable literature on CLC (10-35). Although compared with technologies such as oxy-fuel combustion, CLC offers benefits [1-2], its main limitation is that the CLC process is currently suitable for gaseous fuels only. The wider acceptance of the CLC hinges upon extending the concept to solid fuels [10-11]. The viability of the CLC concept from both technical and economic points of view, especially for future applications involving solid fuels, is underpinned by the availability of effective and robust oxygen carriers [10].

Despite the recent progress in the field [13, 15-17, 24, 26, 29-31] many issues associated with oxygen carriers remain unresolved, particularly:

- the deactivation of oxygen carriers due to physical and chemical phenomena, and;

- the lack of carriers with sufficient level of robustness to allow operations over a wider range of conditions in terms of fuel type and operating parameters.

In addition, CLC is not currently studied in research centres or universities in Australia.

Implementation in interconnected fluidized beds

The interconnected fluidized bed concept of Chalmers University of Technology [10] is a typical design and is illustrated in Figure 2. The oxidized carrier leaves the air reactor (1) by entrainment in the gas stream and is subsequently separated with a cyclone (2). The hot flue gas then exits to a boiler or turbine. The oxygen carrier reacts with the fuel in the reduction reactor (3). Water and non-condensable gases (e.g., unburned fuel) are then separated from the CO_2, which is subsequently captured and stored. The unburned fuel is returned to the reduction reactor, except for a small portion which is bled off into the air reactor to prevent accumulation. Fluidized bed pot seals are used to isolate the air and fuel reactors from each other. To date the concept has been tested using a 10kW prototype [31, 32].

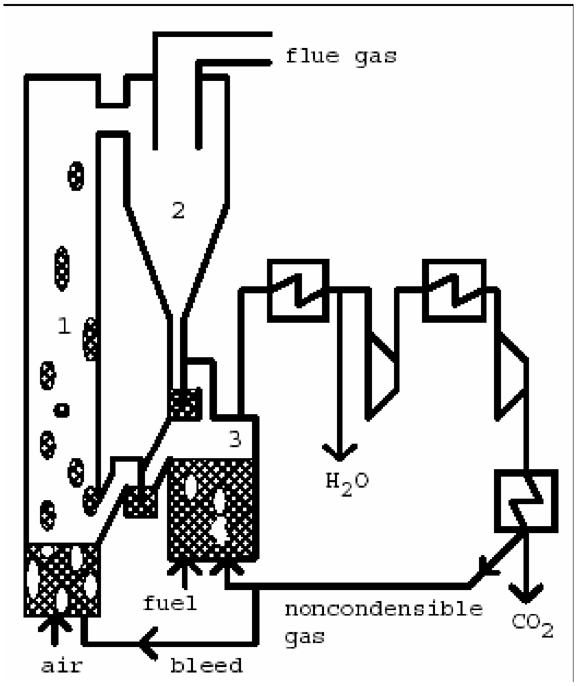

Fig 2: The interconnected fluidised bed design for CLC

Oxygen carriers

Most experimental studies have used Fe_2O_3 as the oxygen carrier although the oxides of Cu, Ni, Mn, and Ba have also been suggested. The fuel oxidation reaction may be endothermic (Fe, Ni) or exothermic (Cu, Mn, Ba) while the solid reoxidation reaction is always exothermic.

Much of the literature on CLC has been devoted to studies concerning metal oxide oxygen carries [13, 15, 22, 31, 29-31]. Successful application of CLC to a great extent relies upon the availability of metal oxides with the following qualities [13]: (i) favourable thermodynamic characteristics to ensure complete fuel conversion, (ii) excellent redox properties in terms of reactivity[1], degree of conversion, selectivity and oxygen transfer capacity[2], (iii) good chemical stability to avoid degradation of redox properties during repeated redox cycles, (iv) high mechanical strength so that particle fragmentation and sintering can be avoided, (v) low cost, and (vi) sound environmental characteristics.

Oxides of transitional metals, such as Fe, Cu, Co, Mn and Ni have been proposed in the literature as potential candidates for large-scale CLC operations. Oxygen carrying characteristics of these metal oxides have been studied under bench-scale experimental settings by several research groups, including: The Tokyo Institute of Technology (Research Laboratory of Resources Utilisation), The National Institute for Resources and Environment (Japan), and The Chalmers University of Technology (Sweden). Bence-scale experiments were typically carried out using temperature programmed thermogravimetric analysers (TGA), pulse reactors, and small-scale fluidised beds. X-ray Diffraction (XRD) and scanning electron microscopy (SEM) techniques were also employed to characterise the surface and physical properties of carrier particles gaining information on both chemical and physical transformations during the redox process [27, 29-31].

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[1] *High reactivity guarantees faster reactions, hence, lowers the quantity of oxygen carrier particles required.*

[2] *A high oxygen transfer capacity means smaller particles can be used. These are much easier to circulate between the two reactors.*

Bench-scale studies have revealed that pure carrier particles are unsuitable for extended use in CLC systems, primarily because of their poor reactivity, chemical stability, and mechanical durability. Inert support materials, such as silica, alumina, yttria-stabilised zirconia (YSZ), kaolin and various metal aluminates were used in past investigations to enhance the mechanical strength and reactivity of carrier particles by improving particle porosity and surface characteristics.

Literature indicates that Fe-based carriers exhibit weak redox characteristics, particularly with respect to reaction rates, even when they are mixed with inert support materials [13, 25, 29]. Also, of the multiple oxidation states of Fe, only the transition between Fe_2O_3 and Fe_3O_4 is thermodynamically favourable for fuel conversion to CO_2 and H_2O. The use of lower oxidation species, such as FeO and Fe, shifts the selectivity towards the formation of CO and H_2 which is not desirable in combustion applications. Furthermore, Fe-based carriers are very susceptible to sintering and formation of agglomerates at temperatures in excess of 800°C. However, because of low cost and environmental compatibility, Fe-based oxygen carries are still considered attractive options for CLC applications [25].

Investigations of Cu-based oxygen carriers have shown that they can achieve higher reduction rate than Fe-based carriers. Lyngfelt and co-workers [13] for instance reported maximum rates of 8%/min and 5.2%/min for reduction of Cu- and Fe-based carriers in the presence of methane (CH_4) at a temperature of 850°C. Unlike other carriers, a notable characteristic of Cu-based carriers is that their reduction is always exothermic [13, 19]. As such, the reduction rate is not limited by the supply of heat from the fuel entering the fuel reactor. The impact of preparation method on chemical and mechanical stabilities of Cu-based carriers has been also investigated in the past [14]. Samples prepared by mechanical mixing, co-precipitation, and wet impregnation were all found to have much higher chemical stabilities than pure carriers under repeated redox cycles. With the exception of samples prepared by impregnation, Cu-based carriers exhibited relatively strong sintering tendencies at temperatures in excess of 800°C [15].

Co-based carriers have generally showed limited conversion capability [10]. It has also been demonstrated that redox properties of Co-based carriers progressively deteriorate throughout the cyclic operation [1] for chemical looping combustion of methane. This has been attributed to chemical instability of Co on alumina support due to the occurrence of secondary reactions between carrier particles and the alumina. However, the use of YSZ support in conjunction with composite CoO-NiO carrier for CLC with coal gas (essentially H_2 and CO) did not show any sign of chemical instability.

The extent of reduction for Mn-based carriers have been found to be quite low with typical reduction rates of approximately 22% of theoretical values and conversion levels of 70% at the end of reduction. Similar to Co-based metal oxides, Mn-based carriers on alumina support have also exhibited [10, 25] chemical instabilities due to reactions between the metal carrier and alumina support to form aluminate ($MnAl_2O_4$). The extent of reactions between Mn-based carries and YSZ support has been found to be insignificant [35].

Compared to other transitional metal oxides, Ni-based carries have been studied in greater detail because of their more favourable characteristics for chemical looping combustion [10-11, 15, 20-22, 30-33]. Ni-based carriers possess good redox, thermodynamic and mechanical properties and relatively high chemical stability. They also seem to have the highest reduction rate when compared with other transitional metal oxygen carriers. Bench-scale results indicate Ni-based carriers are quite suitable for full combustion of methane and its partial oxidation reforming at temperatures between 850^{o}-$950^{o}C$ [1]. No difficulties with sintering, regeneration or mechanical durability have been reported at this range of temperatures. Pilot-scale tests in a 10 kW CLC system indicated a useful lifetime of about 40,000 h for Ni-based carriers [10]. Both alumina and YSZ supports enhance the redox properties of Ni-based carriers although limited reactions between Ni and alumina has been reported [10, 13].

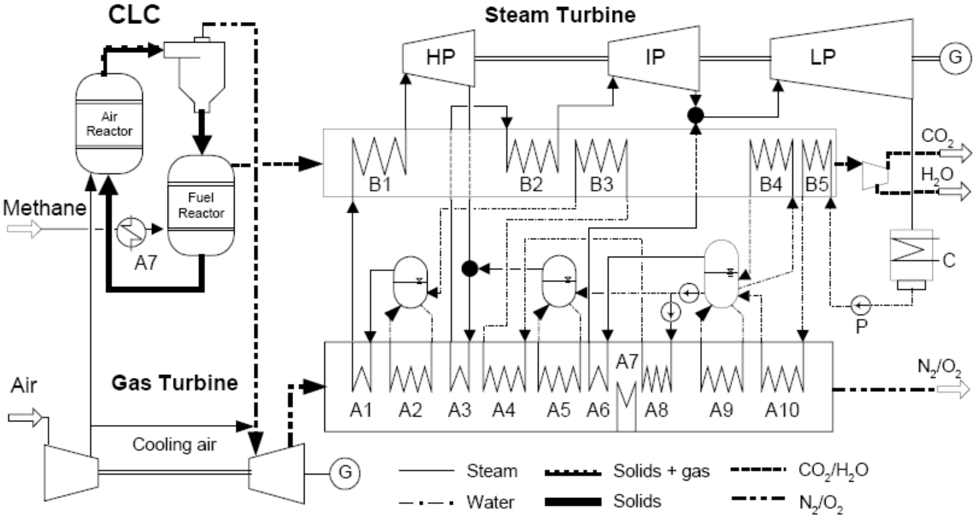
The shortcomings of Ni-based oxygen carriers are that nickel is relatively expensive and quite hazardous. But perhaps the most important drawback of Ni-based carriers is their thermodynamic limitation for full conversion of hydrocarbon fuels (e.g. CH_4) which leads to the formation of undesirable products, such as CO and H_2 [13, 31]. There is, however, inconclusive information about the extent of undesirable by-product formation. Cho and co-workers, for instance [4], reported of only small amounts of CO and H_2 when NiO/Al_2O_3 was reduced in the presence of CH_4 at a temperature of 850^oC. Conversely, Villa et al [31] observed that that CO and H_2 were by far the most abundant products of CH_4 oxidation using a Ni-based oxygen carrier doped with Mg. The greater selectivity of the carrier to CO and H_2 formation in this case was attributed to the impact of Mg on limiting the crystal size of NiO [31].

A common problem with all transitional metal oxygen carriers is the formation of carbon deposits (i.e. coke) on the surfaces of carrier particles during the reduction phase (i.e. fuel oxidation). Ishida and Jin [30] reported that carbon deposits cause degradation of the physical strength of particles and their chemical stability. It has been demonstrated [30-31] that coke formation can be avoided by co-feeding fuel with steam at a molar ratio of 1/1. However, this may detrimentally affect the selectivity of the oxygen carrier leading to formation of more undesirable CO and H_2 by-products. The necessary conditions for carbon deposition on Ni-based oxygen carrier particle were mapped by Ryu [35] and Ishida [30] over a temperature range of $800\text{-}1200^oC$. No coke formation was observed for temperatures greater than 950^oC.

Assessments

In conventional CLC the bulk of the power is generated by the hot flue gas from the air reactor entering a gas turbine (37). Hence, the plant's overall thermal efficiency is highly dependent on the maximum temperature that can be tolerated by the carrier during oxidation for extended periods. Sensitivity analysis shows that thermal efficiency varies from 45% (LHV) at an oxidation reactor outlet temperature of 900°C to more than 55% (LHV) at a temperature of 1200°C. To achieve the efficiencies possible for current gas turbines therefore requires appropriate carriers.

Wolf (38) considered a natural gas-fired combined cycle (NGCC). The potential efficiency of such a process using a turbine inlet temperature of 1200 °C and a pressure ratio of 13 is between 52 and 53 % (LHV) when including the penalty for CO_2 compression to 110 bar.



A1, A3, A6 = superhpeater; A2, A5, A9 = evaporator; A4, A7, A8, A10 = preheater
B1, B2 = superheater; B3 economizer; B4 = supplemental evaporator; B5 = preheater
P = pump; C = condenser; G = generator

Fig 3: Conventional CLC flowsheet used by Wolf (37) with air cooled gas turbine, and steam turbine, incorporating CO_2 generation suitable for capture

Wolf also concluded that co-generation of electrical power and hydrogen is a promising new application for CLC, even if the temperature in the air reactor cannot be raised over 1050 °C. The produced hydrogen can be traded as a valuable byproduct or can be used in the process itself for top-firing.

The potential of chemical looping combustion is illustrated by the results of two simulation studies. In the first [39], HYSIS was used to simulate power generation using an atmospheric pressure fluidized combustion process using natural gas that was modified to incorporate a BaO_2-BaO sorbent . Energy released by the chemical looping reactions was used to generate supercritical steam to produce 500 MWe of electrical power. CO_2 capture approached 100% and the thermodynamic efficiency of the process was 45.5% (LHV). In the second study [40] Aspen Plus was used to simulate a 250 MWe natural gas combined cycle power plant based using a mixed Fe_2O_3-CuO oxygen carrier. Process conditions were chosen so that both reactor effluent temperatures were limited to 900°C to operate with currently available high temperature gas-solid filters and to minimize solid sintering. This case study resulted in a 50% CO_2 capture for a flowsheet designed for high efficiency with an overall plant efficiency of 53% (LHV), associated with an energy penalty of 6% compared to the normal operation of this combined cycle plant.

These examples illustrate that assessments differ, due to CLC flowsheets being quite variable and perhaps based on projected future developments in gas cleaning or turbine capabilities, and that there are trade offs between efficiency, CO_2 capture and oxygen carrier properties.

Developments

Recent CLC research has considered natural gas as the fuel, but Stromberg [41] has stated that CLC should not be discounted in the future for coal firing. Research has recently investigated chemical –looping reforming as a novel procedure for partial oxidation. The process study considered H_2 production through partial reforming of methane in CLC revealed overall efficiency of 81% with possible CO_2 sequestration. The study indicated that a fuel reactor temperature of 1000°C and air preheated temperature of 600°C with system pressure up to 10 bars may yield an overall efficiency of 81% excluding heat losses but including CO_2 capture at 100 bars.It also suggested increasing the air preheated temperature to 650°C and fuel temperature to 1060°C along with reactor pressure up to 16 bars will increase the efficiency by 1% and make the system self sustainable [42]. A further study aimed at achieving higher efficiency in a chemical looping combustion scheme with natural gas combined cycle by introducing reheat in the air turbine through multiple CLC reactors .It was concluded that single reheat combined cycle was preferred as it gave better plant efficiency at safer temperature and had less reactors compared to double reheat combined cycle [43]. Other studies have considered gas turbine combustors based on chemical looping [44].

A further development is that the 2nd Workshop of the Oxy-Fuel Combustion Network organised by IEA Greenhouse Gas R&D Programme and held in January 2007 will have a session on CLC [45]. This will be attended and reported.

Conclusions

Chemical looping combustion enables CO_2 capture without the high efficiency loss of current carbon captures technologies. CLC avoids the need for oxygen production by an ASU, which is a unit operation of the current schemes for pre-combustion and oxyfuel technologies associated with high energy penalties. CLC can be therefore seen as part of a family of technologies which includes membranes which, at present, are progressing through development based on gas fuels, with expectations to application with coal. Electricity generation efficiencies greater than 50% are estimated for CLC, when CO_2 suitable for compression is generated. CLC is more tolerant of the impurities associated with coal combustion than membrane systems, and could be therefore favoured. In addition, the technology uses a fluidised bed technology which is familiar to the combustion engineer, even though this technology is not common in Australia.

The estimated efficiency of CLC using a gas turbine inlet temperature of 1200ºC has been estimated to be between 52 and 53 % (LHV) which includes the penalty for CO_2 compression to supercritical state.

Research has been undertaken primarily in Europe, but with emerging activity now in other areas. Experimental systems have been developed for gas fuels, coal-based CLC is now being developed. The process has been tested in a gas fired 10kw prototype with semi-industrial scale demonstration (> 1 MW_{th}) of CLC being the next step in demonstration. Research has concentrated on system flowsheet assessments for efficiency optimisation and development of robust oxygen carriers.

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