CHALMERS
UNIVERSITY OF TECHNOLOGY

Chemical-looping combustion of solid fuels - Technology overview and recent operational results in 100 kW unit

Downloaded from: https://research.chalmers.se, 2025-12-04 20:21 UTC

Citation for the original published paper (version of record):
Lyngfelt, A., Linderholm, C. (2014). Chemical-looping combustion of solid fuels - Technology overview and recent operational results
in 100 kW unit. Energy Procedia, 63: 98-112. http://dx.doi.org/10.1016/j.egypro.2014.11.011

N.B. When citing this work, cite the original published paper.

research.chalmers.se offers the possibility of retrieving research publications produced at Chalmers University of Technology. It covers all kind of research output: articles, dissertations, conference papers, reports etc. since 2004. research.chalmers.se is administrated and maintained by Chalmers Library

(article starts on next page)

Available online at www.sciencedirect.com

ScienceDirect

Energy Procedia 63 (2014) 98 – 112

Energy Procedia

GHGT-12

Chemical-Looping Combustion of Solid Fuels – Technology Overview and Recent Operational Results in 100 kW Unit

Anders Lyngfelt* and Carl Linderholm

Department of Energy and Environment, Chalmers University of Technology, S-412 96 Göteborg, Sweden

Abstract

Chemical-looping combustion is a new combustion principle that uses metal oxides for oxygen transfer from air to fuel. Fuel is never mixed with combustion air and the combustion products, CO_2 and steam, are recovered in a separate flow without gas separation. Thus, CO_2 capture is inherent in the process and costs and energy penalties for gas separation are avoided. The paper includes:

- an overview of results from a 100-kW chemical-looping combustor for solid fuel.
- a discussion of technology scale-up and costs based on comparison to conventional circulating fluidized-bed boilers.

The added cost for a CLC power plant, compared with a conventional CFB plant, should be in the range 12-27 €/tonne of CO_2 avoided, and likely around 19-20 €/tonne.

© 2014 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/3.0/).
Peer-review under responsibility of the Organizing Committee of GHGT-12

Keywords: Chemical-looping combustion; oxygen carrier; solid fuels, circulating fluidized bed; CO_2 avoided cost

* Corresponding author. Tel.: +46-31-772-1427
E-mail address: Anders.Lyngfelt@chalmers.se

1876-6102 © 2014 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/3.0/).
Peer-review under responsibility of the Organizing Committee of GHGT-12
doi:10.1016/j.egypro.2014.11.011

1. Introduction

Chemical-looping combustion (CLC) has emerged as an attractive option for carbon dioxide capture because CO_2 is inherently separated from the other flue gas components, i.e. N_2 and unused O_2, and thus no energy is expended for the gas separation and no gas separation equipment is needed. The CLC system is composed of two interconnected fluidized bed reactors, an air and a fuel reactor, [1]. Oxygen carriers in the form of metal oxide particles are used to transfer oxygen between the two reactors, Fig. 1.

CLC research has mainly focused on gaseous fuels, but in the last years important work has been dedicated to adapting the process to solid fuels. For more detail a number of reviews are available, [2-8]. First to study solid fuels for CLC was Lewis et al. [9], using copper and iron oxides and fifty years later a number of new laboratory studies emerged, involving the same oxides as well as manganese and nickel, see e.g. [4]. Today there are a number of publications of actual operation in smaller CLC pilots with solid fuel, e.g. [10-19].

The direct use of solid fuels in CLC could use the circulating fluidized bed (CFB) concept outlined in Fig. 2, but the fuel reactor system would need to be adapted for addition of solid fuels. When using solid fuels, the reaction between the oxygen-carrier and the char remaining after volatiles release is not direct, but involves an intermediate gasification step, Fig. 3. This is determinant for the fuel reactor design and the following key targets have been identified for fuel reactor performance:

- High solid fuel conversion, i.e. minimize loss of unconverted char with flue gas
- High gas conversion, i.e. minimize unconverted gases like H_2, CO and CH_4
- High CO_2 capture, i.e. minimize loss of char to air reactor
-

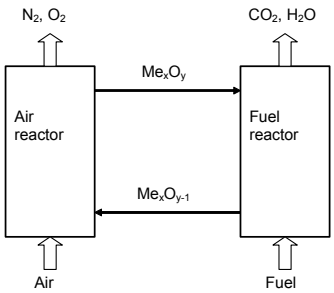

Fig. 1. CLC principle

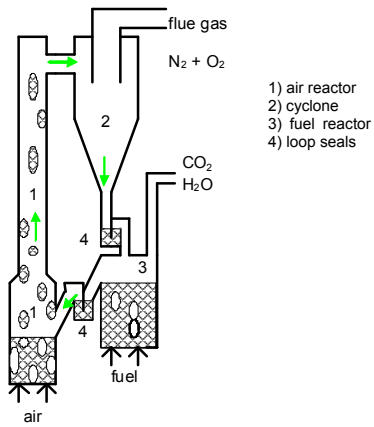

Fig. 2. CFB reactor system for gas

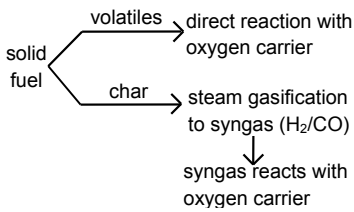

Fig. 3. Sold fuel reactions in CLC

Consequently the performance of operation is estimated based on three performance indicators:

i. Fuel conversion, η_F, is the ratio of carbon converted to gaseous compounds in fuel and air reactor to total carbon added. Thus, $1-\eta_F$, is the fraction of total carbon added that is elutriated from the fuel reactor in the form of char particles.

ii. Oxygen demand, Ω_{OD}, is the fraction of oxygen lacking to fully oxidize the gas from the fuel reactor related to the total oxygen needed to oxidize the same gas. $1-\Omega_{OD}$ is the gas conversion. The oxygen demand gives an indication of the oxygen needed for the oxy-polishing.

iii. CO_2 capture, η_{CO2}, is the fraction of gaseous carbon that leaves the fuel reactor related to the total carbon converted to gas in fuel and air reactors. Thus, $1-\eta_{CO2}$, is the ratio of the carbon that is lost as CO_2 from the air reactor to the total carbon in gaseous compounds leaving air and fuel reactors.

As compared to gaseous fuels, CLC with solid fuels will require a different design of the fuel reactor, and likely also oxygen carriers with other properties:

- The ash, normally being part of solid fuels, will make a very long lifetime of the oxygen carrier unlikely, as the ash removal inevitably causes losses of oxygen carrier. Also, the ash may affect the oxygen carrier directly. This means that the oxygen carrier should have low cost.
- The gasification of char is a slow process, which means that the fuel reactor needs a design that provides sufficient residence time, in order to avoid char particles reaching the air reactor. Char burning in the air reactor should be avoided, as it will produce CO_2 that will not be captured.

In order to achieve high conversion of the volatiles, the fuel needs to be fed to the fuel reactor in a way that allows good contact between the volatiles released and the bed material consisting of oxygen carrier.

An advantage for CLC with solid fuels is that most oxygen carriers, including low-cost materials, are highly reactive towards the syngas released. However, the syngas is released from char particles inside the fuel reactor, in contrast to gaseous fuels which are introduced from below. Thus, some of the syngas released, e.g. in the upper regions, will have insufficient contact with the bed material. Thus, complete conversion of the gas is difficult to obtain. Measures to reach complete or very high conversion include:

- Introduction of pure oxygen downstream of the fuel reactor, in order to oxidize the remaining unconverted gases H_2, CO and CH_4, so-called ‘‘oxygen polishing”.
- The separation of these unconverted gases from the CO_2 in connection with CO_2 liquefaction, followed by recirculation of these gases to the fuel reactor.
- Two fuel reactors in series, i.e. leading the incompletely converted gas from the first fuel reactor to a second fuel reactor.
- Use of a CLOU oxygen-carrier, [20], i.e. a material able to release oxygen in the fuel reactor

1.1. Cost and energy penalties

CLC of solid fuels clearly has a potential for a dramatic reduction of energy penalty and costs for CO_2 capture. Thus, the energy penalty for chemical-looping combustion would ideally be equal to the power needed for CO_2 compression of around 2.5%-units.

A power plant using solid fuel CLC would have significant similarities to a CFB power plant, which is a commercial technology for plants up to 600 MWe. The air reactor would be very similar to a CFB, with some notable differences, such as the need for higher solids circulation, and a smaller gas flow as the oxygen is consumed and no combustion products are released. The gas flow through the fuel reactor is the flow that is not going through the air reactor, i.e the combustion products CO_2 and steam, typically 20-25% of the total gas flow, plus the extra flow of gas for fluidization. Thus, the fuel reactor should be considerably smaller than the air reactor. The fuel reactor is also adiabatic. Clearly, the fuel reactor will add to the cost of the boiler, but on the other hand the higher temperature in the air reactor will mean a reduction in the heat transfer surface areas which will give a cost reduction. Nevertheless, the similarities would be significant, and the cost of the boiler system is typically 30-40% of the total cost of a power plant. So, in all, the additional costs for such an ideal CLC system would be expected to be moderate in comparison to other CO_2 capture technologies.

In the EU project ENCAP a first design of a 455 MWe CLC solid fuel power plant was made. A comparison to a similar fluidized bed combustion power plant indicated a very low efficiency penalty, <3%, as well as a very low capture cost, 10 €/tonne of CO_2, [21]. Later, the European Technology Platform for Zero Emission Fossil Fuel Power Plants estimated the cost for CLC to be 25.7 €/tonne CO_2 avoided, again significantly lower than any other technology, [22].

An advantage with CLC is that the gas coming out from the air reactor can be expected to be essentially free from harmful emissions such as NO_x and SO_2. Compounds formed from nitrogen and sulphur in the fuel will be concentrated in the smaller gas stream from the fuel reactor, which should facilitate separation. Although more development work is needed, especially with respect to finding the best design of the fuel reactor system, it is clear that the CLC technology provides a unique potential for avoiding the large costs and energy penalties inherent in gas separation.

However, little detail on the basis for the cost estimations for CLC has been published, so it is not clear what assumptions have been made and how well these correspond with the present status of the technology development.

1.2. Scope

The paper gives an overview of, and comment to, the operational experiences of operation of a 100 kW unit. Details are given by Markström et al. [23-26] and Linderholm et al. [27-29]. Moreover, the design and cost of a 1000 MW_{th} CLC power plant, i.e. approximately 410 MW_e, are discussed based on a work by Lyngfelt and Leckner, [30].

2. The 100 kW chemical-looping combustor

The system includes two interconnected circulating fluidised beds, i.e. the air and the fuel reactor, as well as a carbon stripper. Starting in the fuel reactor, the path of solids circulation is outlined below and is numbered from 1–28 in Fig. 4.

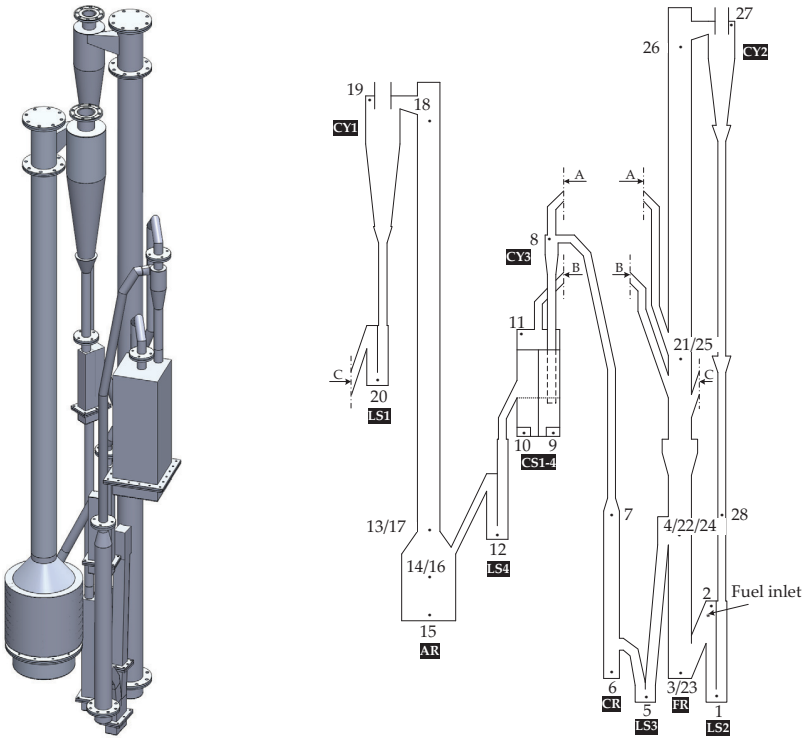

Fig. 4. 100 kW CLC for solid fuels. Left: 3-D representation; Right: 2-D representation;

Gas and particles entrained in the fuel reactor (FR) enter the cyclone (CY2), fall down the downcomer to a loop seal (LS2) and enter the fuel reactor again. Particles not entrained may instead enter the so-called circulation riser (CR), which is placed in between the carbon stripper (CS1-4) and the loop seal (LS3) connected to the fuel reactor. The circulation riser is designed to return varying mass flows of particles to the air reactor. The carbon stripper has four chambers separated by weirs. The purpose of the carbon stripper is to gasify or separate residual char in the particle flow. The particles in the carbon stripper are passed on to a loop seal (LS4), leading to the air reactor (AR). The air reactor will re-oxidise the oxygen carriers before they are transported back to the fuel reactor by passing a cyclone (CY1) and another loop seal (LS1), beginning a new cycle. The air and fuel reactors are separated by loop seals in order to avoid gas mixing. The fuel reactor has an inner height of 5.0 m. The fluid dynamics of the system have been investigated in a cold-flow model [31]. A film of the model in operation is available on http://www.entek.chalmers.se/~anly/co2/CFM.htm

3. Results from 100 kW pilot

3.1. Operation with bituminous coal and ilmenite

The 100 kW unit has been in operation for totally 116 h with different fuels, different oxygen carriers and under different operational conditions.

The first operation reported by Markström et al. [23-26], involves mainly operation with ilmenite and a bituminous coal. The oxygen demand was found to be strongly dependent on the solids inventory, whereas no correlation to solids circulation was seen within the ranges studied. Lowest oxygen demand was 16%.

Operational data of oxygen demand sampled after stops of fuel addition, when volatiles release is effectively stopped, but the release of syn-gas from char continues, clearly indicate much lower oxygen demand for the syn-gas. This is clearly seen in the summary of the results in Fig. 5, where gas conversion data for coal are shown as circles and data for char is shown as squares and diamonds. The two curves are the model results for bituminous coal (lower curve) and char only (upper curve). These results are discussed in the following section.

The much poorer gas conversion for the volatiles was also shown by a short period where steam fluidization failed and the fuel reactor was instead fluidized by N_2, [31]. The fluidization with N_2 can be expected to reduce the rate of syn-gas release substantially, whereas the release of volatiles is unaffected. Thus, the increase in oxygen demand during this period can be explained by the poorer conversion for the volatiles.

3.2. Gas by-pass and model results

An analytical model was used to analyze the results, and good fit between experimental results and model was reached when the parameters describing the contact of gas and solids, i.e. $\phi_{s,core,v}$ and $\phi_{s,core,c}$ were given the values 0.15 and 0.9. $\phi_{s,core}$ is defined as the fraction of solids, i.e. oxygen carrier, that is exposed to the gas flow which is assumed to move in plug flow. The model has two values for this parameter, one for the volatiles, $\phi_{s,core,v}$, and one for the syn-gas released from the char, $\phi_{s,core,c}$.

Starting with the low value for the volatiles, 0.15, it clearly illustrates a poor contact between gas and solids, which is also typical and expected for the fluidizing conditions in this fuel reactor. According to the two-phase theory of fluidization, only a small fraction will move upwards inside the dense particle phase, whereas the majority of the gas will travel upwards in a dilute phase at high velocity. Furthermore, the ratio of bed height to bed diameter is high, and therefore slugging conditions would be expected, which is also the case as shown by the large pressure fluctuations measured. The same value of $\phi_{s,core,v}$, was previously also to model a chemical-looping combustor of similar size using gaseous fuel, [32].

The high value of the same parameter for the syngas can be explained as follows. Whereas the volatiles are released in the freeboard of the loopseal exit as the fuel is introduced and will become part of the dilute phase with poor contact with the solids, this is not the case for the char. The char remaining after the volatiles release can be expected to fall into the dense phase and mix with the oxygen carrier. This means that the syn-gas is to a large extent released inside the dense phase in close contact with the oxygen-carrier material. Furthermore, the syn-gas released, becomes highly concentrated as it is not diluted with the large flow in the dilute phase. The higher concentration is synonymous with higher conversion rates as these reactions are first-order or close to first-order. This also explains why the conversion of syngas is very much higher than that of the volatiles.

The model assumes that the volatiles are introduced in the bottom of the reactor, whereas the syngas release is distributed equally over the bed. Generally, this should give a significant disadvantage for the syngas conversion, and the model results by Abad et al., [33], came to the opposite conclusion that gas conversion for the volatiles was significantly better as compared to the syngas from the char.

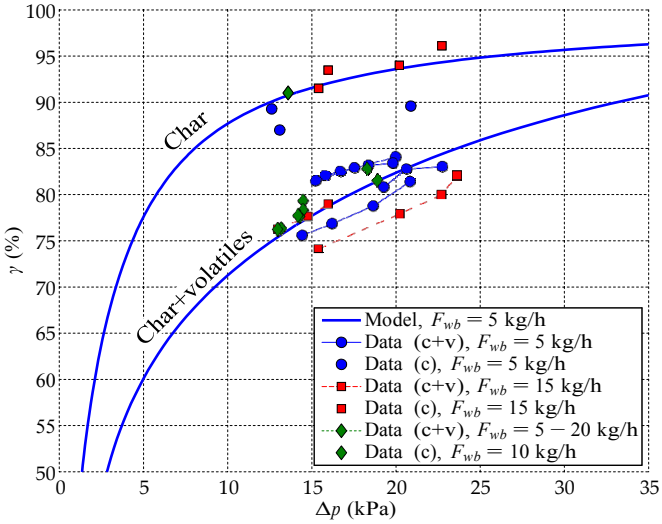

Fig. 5. Gas conversion in 100 kW versus solids inventory for ilmenite using coal. c and v denote char and volatiles.

3.3. Operation with low-volatile fuel and ilmenite

Later operation with a low volatile fuel, Swedish wood char, also clearly indicates the higher gas conversion associated with the syn-gas. Fig. 6 also shows flows, global circulation, pressure drop in FR, oxygen demand and carbon capture efficiency as functions of time for 4.9 h of operation under very stable conditions. The oxygen demand is typically 5% and the lowest value is 3.7%. The minimum oxygen demand, 1.97% is observed right after fuel termination, after 5.05 h.

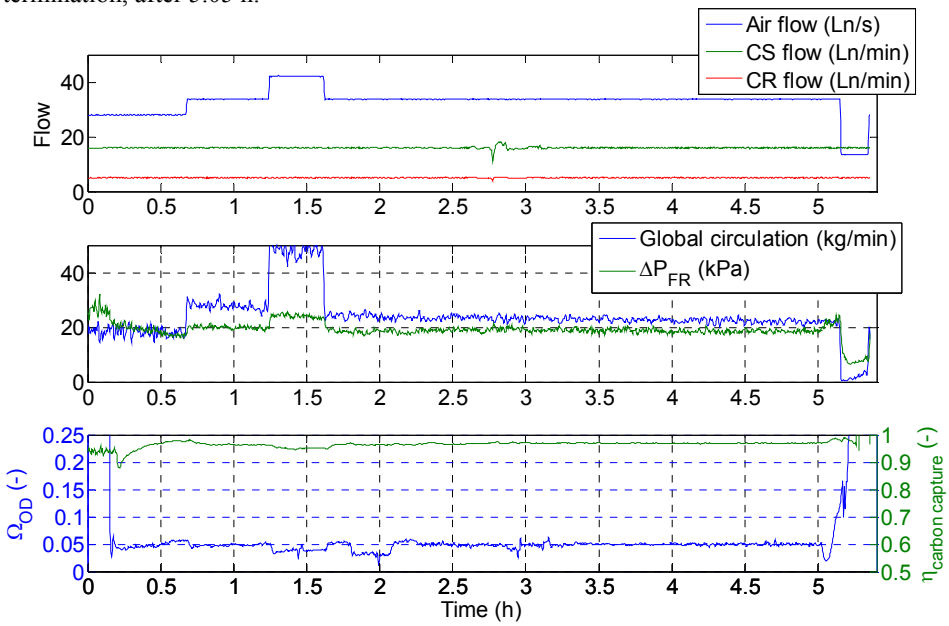

Fig. 6. Flows, global circulation, pressure drop in FR, oxygen demand and carbon capture efficiency as functions of time. For twenty minutes around 2 h, nitrogen is replaced with argon. The low oxygen demand seen in this period is an artefact.

3.4. Operation with ilmenite and addition of manganese ore

During three days a total of 18 hours of operation with ilmenite with addition of manganese ore was achieved. The results are shown in Fig. 7, together with reference data for ilmenite only. The fraction of manganese ore can be expected to increase during the three days of operation, which is also reflected in the increased performance.

Because of higher elutriation of the manganese ore, the fraction of manganese ore in the bed material was still low at the end of the test series, around 10% according to post-operational analysis of bed samples. Nevertheless, a dramatic increase in the gas conversion was seen, and the lowest oxygen demand was 8.5% to be compared to the best for ilmenite with the same coal, 16%. Thus, it is clear that significantly improved gas conversion is possible with manganese ore.

Furthermore, the loss of unconverted char is also reduced when manganese ore is added. This effect has been noted previously, [15], and is explained by the presence of some sodium and potassium in the manganese ore, [34, 35].

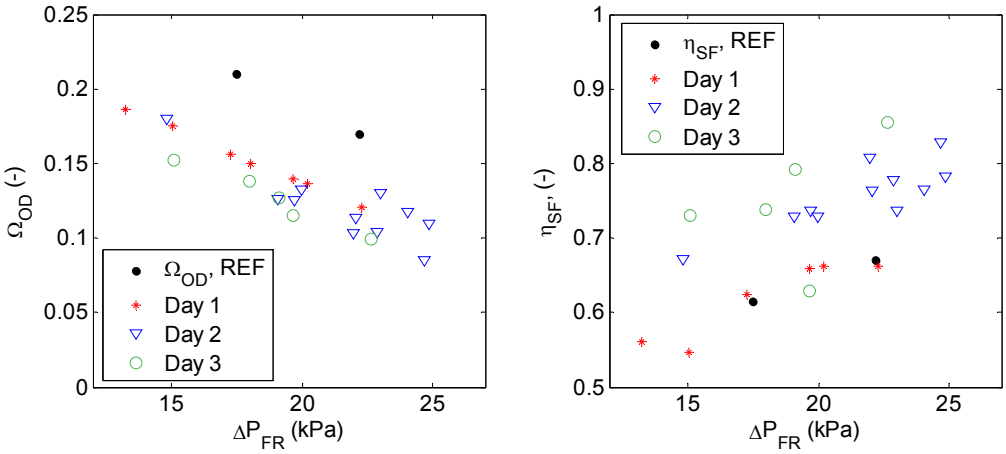

Fig. 7. Oxygen demand (left) and solid fuel conversion (right) as functions of the fuel-reactor pressure drop. Red and blue markers: Cerrejon coal. Green markers: Polish coal. Black markers indicate reference tests performed Day 1 with only ilmenite oxygen carrier.

3.5. Implications for design of operational experiences

The operational results in the 100 kW unit clearly indicated the benefits of a high solids inventory. Furthermore, it is also very important to design a fuel reactor for good contact between oxygen carrier and volatile gases.

4. 1000 MW_{th} design

A tentative design of a 1000 MW_{th} CLC power plant has been proposed, [30], corresponding to a power of approximately 410 MW_e. It shows that such a unit could be reasonably similar to commercial CFB power plants. A CFB power plant with a thermal power of 1000 MW_{th} was used as reference, and a similar height was used for the CLC plant as the ref, i.e. 48 m. The sum of the gas flows is almost the same as in the reference case, but with 27% in the fuel reactor and the remainder in the air reactor. Further, a higher velocity was assumed in the air reactor in order to achieve the circulation needed. A comparison of the reference CFB and the CLC boiler is given in Fig. 8.

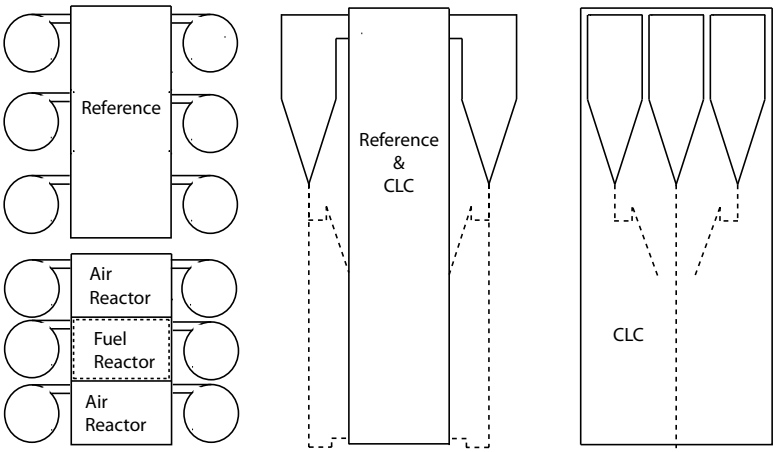

Fig. 8. Top/side view of CFB and CFB-CLC (left/middle), front view CLC (right)

An important difference is that the fuel reactor is adiabatic, which will add an extra cost for insulation. Pilot operation indicates a strong correlation between solids inventory in the fuel reactor and gas conversion. Therefore a solids inventory in the fuel reactor of 500 tonnes was used, corresponding to a pressure drop of 64 kPa or a settled bed height of 3 m. The heat balance indicated that a solids circulation flow of 5.3 tonnes/s is needed to keep the temperature difference between the reactors within 50ºC. With a temperature of 1020ºC in the air reactor this means 970ºC in the fuel reactor.

In the fuel reactor the fluidizing velocity is the same as in the reference plant. For the air reactor, however, it was assumed that higher velocity is needed to reach the required circulation. Therefore the cross-section of the CLC plant is smaller in Fig. 8. The particle flow from the four air reactor cyclones will be led to the fuel reactor, and added through the external walls above the bottom dense region.

The material from the fuel reactor cyclones will contain combustibles in the form of char, and this material will be led to the bottom of the dense region of the fuel reactor bed. The fuel will be fed into this stream and the volatiles released will be led into a volatiles distributor as described in the following section. The bed material is returned from the fuel reactor to the air reactors via a loop seal below the beds, using the same principle as in the Vienna 100 kW CLC, [36]. The loop seal connection is located at the centre line, i.e. at maximum distance from the wall where fuel and circulated oxygen carrier material is introduced.

In order to reach full conversion a post-oxidation chamber for oxy-polishing of the gas leaving the fuel reactor is also needed.

4.1. Volatiles distributor

In order to avoid a local plume of volatiles where fuel is added, or avoiding a large number of fuel entries, a device for distributing fuels is used. The principles of such a distributor are described in a patent application, [37]. It is based on the fact that a box immersed in a fluidized bed with the opening downward will become empty, i.e. the bed surface will follow the lower edge of the box. If such a box has openings or holes on the sides, the bed surface will rise to the level of the openings. In short, the distributor can be described as an elongated box with the opening downwards and holes or slits on the sides. If this distributor is immersed in a fluidized bed, the bed material forms a bed surface at the level of the holes. Above this surface, gas can flow freely. If gas is injected in this distributor it will leave the distributor via the holes. The fuel is added in such a way that the volatiles are released in a gas volume that is connected with the distributor. Furthermore, the distributor will involve a number of ”arms” to assure a good distribution of volatiles over the cross-section.

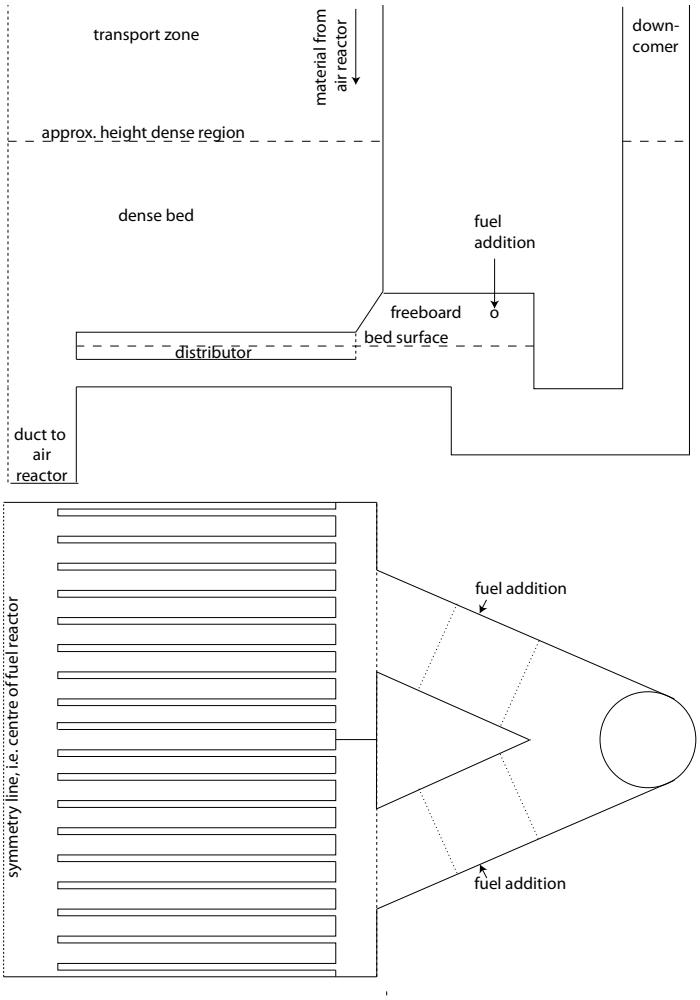

Fig. 9. Side/top view of the right-hand half of the fuel reactor (top/bottom), showing distributor and solids return.

A possible lay-out of such a distributor and its placement in the bottom part of the fuel reactor is given in Fig. 9. The detailed design would need to be optimized using experimental data from cold-flow model testing in combination with fluid-dynamic modelling, as well as cost analysis. Such a construction would likely have limited lifetime considering the conditions in the fuel reactor, making regular replacements necessary. Nevertheless, it is worthwhile considering the importance of good contact between reacting gas and oxygen carrier to achieve high gas conversion and thus to minimize the need for oxygen production.

4.2. Discussion 1000 MW_{th} design

The need to reach the adequate solids circulation is an important restriction on the design. Too low circulation will lead to higher temperature difference between air and fuel reactor, i.e. a lower temperature in the fuel reactor which will slow down the char gasification, thus giving poorer performance with respect to fuel conversion and CO_2

capture. The needed circulation is 5.3 tonne/s, which corresponds to a circulation of 25.5 kg/m^2,s using the same conditions in the air reactor as in the reference plant, i.e. the same cross-section per gas flow or a fluidizing velocity of 5.4 m/s. Normally the circulation in is not known or not published, but there are some data in the literature. According to Yue et al., [38], the measured circulations in large CFBs is around 6-10 kg/m^2,s at fluidizing velocities in the range 5-6 m/s. These numbers are in contrast with the finding of Haibo et al., [39], where the circulation in a 300 MW_e at 5.4 m/s is 42 kg/m^2,s. To be noted is that the needed circulation in the air reactor is approximately an average of these two values.

There are several ways to attain the circulation needed:

- higher velocity, i.e. smaller cross-section of the air reactor
- smaller particles
- lower height of air reactor
- use of a separate, dedicated riser to move particles to the fuel reactor

In the design proposed it was assumed that the circulation needed could be obtained by an increased velocity, but high velocities involve risks of erosion of the walls. Therefore, other options to reach needed circulation could be preferable. An important feature of an air reactor is the absence of fuel producing combustible gases and char that need to be well mixed with the combustion air and have sufficient residence time to become fully converted. This absence of fuel means that there is no need for gas residence time. In other words there is no need for, or advantage of, a high reactor. Moreover, as discussed below, the air reactor will also need less cooling surfaces as compared to the CFB boiler. As internal panel walls or external fluidized heat exchangers can be used to increase heat extraction there is no need of a high reactor for the heat transfer. Thus, the minimum height of the air reactor is determined by the height of the cyclones and the ducts needed to feed oxygen carrier to the fuel reactor. As illustrated by Fig. 10, it would be possible to reduce the height of the air reactor significantly.

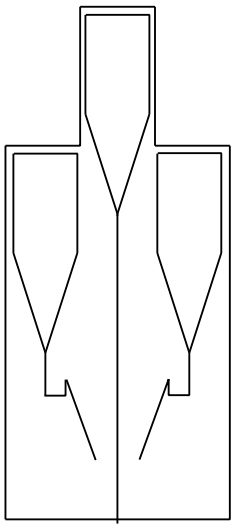

Fig. 10. Side view of CLC with reduced air reactor height.

The effect of reduced height on the solids circulation can be estimated by, K, the decay constant. Johnsson and Leckner proposed an estimation of the decay constant by $K = 0.23/(u-u_t)$ where u is the fluidization velocity and u_t is the terminal velocity, [40]. With $u-u_t$ values in the range 4-5 m/s, the decay constant becomes around 0.05. The corresponding effect of the decrease in height of 13 m, as shown in Fig. 10, is approximately a doubling of the circulation. It is likely possible to reduce the air reactor height further depending on the design and placing of cyclone, loop-seal and ducts. It can be concluded that a reduced air reactor height is an effective measure to increase solids circulation.

For the overall cost calculations the method used to reach adequate circulation only has a small effect.

5. Cost analysis

An estimation of the added cost for a CLC plant was made, [30], based on the differences between a CLC and CFB. The main costs are presented below.

5.1. CO_2 compression and downstream treatment

CO_2 compression and downstream treatment was assumed to give an added cost of 10 €/tonne CO_2 avoided. This is mainly cost for compression. Downstream treatment, e.g. removal of sulphur and nitrogen compounds, was assumed to have little cost because these compounds are concentrated and leave the system in a smaller stream in the CLC case. This should facilitate the removal and therefore it is assumed that this will not give any significant added costs, as compared to the reference case.

5.2. Oxy-polishing

It was assumed that the need for oxygen for oxy-polishing is 5-15% of the oxygen needed to totally burn the fuel. Further the total cost for production of oxygen for the oxy-fuel process was assumed to be 40 €/tonne CO_2 avoided. To take into account that the specific cost for oxygen production falls with scale, it was assumed that the air separation cost is proportional to the amount of oxygen raised by a power of 0.8. This means that the cost would be reduced by 77-90% as compared to oxyfuel, or 4-9 €/tonne CO_2 avoided.

5.3. Boiler cost

As noted above, the fuel reactor needs to be insulated and, in contrast to the corresponding walls of a CFB boiler, these insulated walls will not be involved in the transfer of heat to the steam. This means that this heat transfer duty has to be accomplished elsewhere. For this reason the walls of the fuel reactor are considered to be an extra cost. Furthermore, there is also an added cost of the walls of the oxy-combustor where the oxygen polishing will take place. Moreover, the cost of insulation for the cyclones of the fuel reactor and ducts leading material to the fuel reactor was added. Total added insulated area was estimated to 3200 m^2, corresponding to a cost of 0.2 €/tonne CO2 avoided.

The primary functions of a boiler are to constitute the room where the combustion takes place, to transfer the heat to the steam used to produce power and to avoid loss of heat to the surroundings. Normally, panel walls are used which serve the double purpose of heat transfer and being walls of the combustion chamber. For large boilers with large volume to surface ratio, the heat transfer given by the external walls is insufficient and heat transfer surface need to be added. The heat transfer surfaces are also expensive as they need to operate at high temperatures with the combustion chamber on one side and steam production on the inside. Thus, they constitute an important part of the boiler costs.

In the case of CLC the air reactor will have a much higher temperature than a CFB, and thus much higher heat transfer, which is estimated to reduce the needed heat transfer area by 45-50%. On the other hand the needed heat transfer area in the convection pass is likely not significantly affected. Nevertheless, this could mean that the CLC boiler costs could be significantly reduced. Assuming that the boiler constitutes 33% of the total investment cost and that the boiler cost is changed by maximum ±20%, this would correspond to a change in CO_2 capture cost of ±4 €/tonne CO_2. It can be noted that the cost of the added insulation is small in comparison.

Based on the presently available understanding it appears likely that the cost of the boiler would be lower for the CLC as compared to the CFB reference. However, considering the uncertainties, it is assumed that cost is in the range ±4 €/tonne CO_2.

5.4. Oxygen carrier cost

The cost of the oxygen carrier is 1-3 €/tonne CO_2, assuming a cost of oxygen carrier, i.e. ilmenite, of 100 €/tonne and a lifetime of 100-300 h.

5.5. Penalty for fluidization or air and fuel reactors

The fuel reactor needs to be fluidized with a gas that does not dilute the CO_2. The available options are to use steam, CO2 or "hot" CO_2, the latter meaning gas recycled that is extracted before the steam is removed. Thus, the "hot" CO_2 contains steam which is an advantage for the gasification, as compared to CO_2. Best for the gasification is steam, but steam production has a larger negative effect on the energy penalty. A fluidization gas of 50% steam and 50% "hot" CO_2 was therefore assumed. The associated energy penalty is around 0.8%.

A higher solids inventory in the air reactor will give a higher pressure drop, as compared to the CFB reference, giving an energy penalty for the air fans of around 0.2%. Totally, the penalty for the air and fuel reactor fluidization was estimated to 1%, the major part of this penalty coming from the steam production.

The added cost for this energy penalty is around 1 €/tonne CO_2.

5.6. Summary cost analysis

In total the estimated added costs for a CFB-CLC are in the range 12-27, *cf.* Table 1. The largest cost is that of CO_2 compression which is common for all CO_2 capture technologies. Normally, the CO_2 compression cost is small in comparison to the "capture" cost, i.e. attaining a pure CO_2 to compress. This is not the case for CLC, where the estimated "capture" cost could be as low as 2 €. The largest uncertainties are in the cost for the boiler and for the oxy-polishing. The latter is associated with the uncertainty of the performance and based on an oxygen demand in the range 5-15%. If we assume that the large reduction in heat transfer area for the boiler compensates other added boiler cost, that the gas conversion is 90%, and that the oxygen carrier cost is in the middle of the range, the resulting cost is 19-20 €/tonne of CO_2.

Table 1. Estimated added costs for CFB-CLC, €/tonne CO2 avoided.

Type of cost	estimate, €/tonne CO_2	range, €/tonne CO_2
CO_2 compression	10	10
Oxy-polishing	6-7	4-9
Boiler cost	<0	±4
Oxygen carrier	2	1-3
Steam and hot CO_2 fluidization	1	1
Total	19-20	12-27

6. Summary of results and conclusions

A summary of experimental results is given below. For details see [23-29].

- It is difficult to reach full gas conversion.
- With pulverized fuel it does not seem to be difficult to reach very high CO_2 capture, e.g. >98%.
- Reasonably high gas conversion was reached with bituminous coal, using the low-cost oxygen carrier

ilmenite, at best 84%.

- Gas conversion was highly correlated to solids inventory.
- Much higher gas conversion was reached with low-volatile fuels, e.g. 95% for wood char, clearly indicating that contact between oxygen carrier and volatiles is a major issue for the gas conversion.
- Gas conversion was improved by adding manganese ore to the ilmenite bed material, 91-92% at best, which demonstrates that it is possible to increase conversion further with other oxygen carriers.

The operational experience gives a strong confirmation of the viability of CLC with solid fuels.

The design of a CLC-CFB has been discussed with focus on differences compared to CFB technology and the possible implications with respect to costs. In addition to CO_2 compression, which is inevitable and common to all CO_2 capture technologies, important costs are air separation for oxy-polishing, oxygen carrier and fluidization of fuel reactor. The cost for air separation could be low if gas conversion is high, also the cost of oxygen carrier could be low if lifetime is high. Finally, the boiler cost could be both lower and higher as compared to the reference CFB technology. The cost for CO_2 capture is estimated to be 19-20 €/tonne CO_2 avoided, and should be within the range of 12-27 €/tonne.

7. Acknowledgement

The research leading to these results has received funding from the European Council under the European Union's Seventh Framework Programme (FP7/2007-2013) ERC grant agreement no 291235.

The experimental work referred to was carried out with financial grant from the Research Fund for Coal and Steel of the European Community (ECLAIR project, contract RFCP-CT-2008-0008 and ACCLAIM project, contract RFCP-CT-2012-0006) and Alstom Power Boilers.

8. References

[1] Lyngfelt, A., Leckner, B., and Mattisson, T., (2001), A fluidized-bed combustion process with inherent CO_2 separation; Application of chemical-looping combustion, Chemical Engineering Science, 56(10), pp. 3101-3113.

[2] Hossain, M.M. and de Lasa, H.I., (2008), Chemical-looping combustion (CLC) for inherent CO2 separations-a review, Chemical Engineering Science, 63(18), pp. 4433-4451.

[3] Lyngfelt, A., (2011), Oxygen carriers for chemical-looping combustion - 4000 h of operational experience, *Oil & Gas Science and Technology - Revue d'IFP Energies nouvelles* 66(2), pp. 161-172.

[4] Lyngfelt, A. and Mattisson, T., *Materials for chemical-looping combustion*, in *Efficient Carbon Capture for Coal Power Plants*, D. Stolten and V. Scherer, Editors. 2011, WILEY-VCH Verlag GmbH & Co.. KGaA: Weinheim. p. 475-504.

[5] Adánez, J., Abad, A., Garcia-Labiano, F., Gayan, P., and de Diego, L., (2012), Progress in Chemical-Looping Combustion and Reforming technologies, Progress in Energy and Combustion Science, 38(2), pp. 215–282.

[6] Linderholm, C., and Lyngfelt, A., , *Use of chemical looping combustion for solid fuels*, in *to be published as Chapter 14 in Calcium and chemical looping technology for power generation and carbon dioxide (CO2) capture*, P. Fennell and E.J. Anthony, Editors. 2015, Woodhead Publishing.

[7] Lyngfelt, A., *Chemical looping combustion*, in *Fluidized-bed technologies for near-zero emission combustion and gasification*. 2013, Woodhead Publishing Limited. p. 895-930.

[8] Lyngfelt, A., (2014), Chemical-looping combustion of solid fuels – status of development, Applied Energy, 113, pp. 1869-1873.

[9] Lewis, W., Gilliland, E., and Sweeney, M., (1951), Gasification of carbon Chemical Engineering Progress, 47(5), pp. 251-256.

[10] Berguerand, N. and Lyngfelt, A., (2008), Design and operation of a 10 kWth chemical-looping combustor for solid fuels - Testing with South African coal, Fuel, 87(12), pp. 2713-2726.

[11] Berguerand, N. and Lyngfelt, A., (2008), The use of petroleum coke as fuel in a 10 kWth chemical-looping combustor, International Journal of Greenhouse Gas Control, 2(2), pp. 169-179.

[12] Shen, L., Wu, J., and Xiao, J., (2009), Experiments on chemical looping combustion of coal with a NiO based oxygen carrier, Combustion and Flame, 156(3), pp. 721-728.

[13] Cuadrat, A., Abad, A., García-Labiano, F., Gayán, P., de Diego, L.F., and Adánez, J., (2011), The use of ilmenite as oxygen-carrier in a 500Wth Chemical-Looping Coal Combustion unit, International Journal of Greenhouse Gas Control, 5(6), pp. 1630-1642

[14] Wu, J., Shen, L., Hao, J., and Gu, H., *Chemical looping combustion of coal in a 1 kWth reactor*, in *1st International Conference on Chemical Looping*. 2010: Lyon.

[15] Linderholm, C., Lyngfelt, A., Cuadrat, A., and Jerndal, E., (2012), Chemical-looping combustion of solid fuels – operation in 10 kW unit with two fuels, above-bed and in-bed fuel feed and two oxygen carriers, manganese ore and ilmenite, *Fuel*, 102, pp. 808–822.

[16] Sozinho T, Pelletant W, Gauthier T, and Stainton H, *Main results of the 10 kW coal pilot plant operation.* , in *2nd Int. Conf. on Chemical Looping*. 2012: Darmstadt.

[17] Thon A, Kramp M, Hartge EU, Heinrich S, and Werther J, (2014), Operational experience with a system of coupled fluidized beds for chemical looping combustion of solid fuels using ilmenite as oxygen carrier Applied Energy, Volume 118, pp. 309–317.

[18] Tong, A., Bayham, S., Kathe, M., Zeng, L., Luo, S., and Fan, L.-S., *Iron-Based Syngas Chemical Looping Process and Coal-Direct Chemical Looping Process Development at Ohio State University*, in *2nd International Conference on Chemical Looping*. 2012: Darmstadt.

[19] Ströhle, J., Orth, M., and Epple, B. (2013) *Chemical Looping Combustion of Hard Coal in a 1 MW_{th} Pilot Plant Using Ilmenite as Oxygen Carrier*. in *3rd International Conference on Chemical Looping*. Göteborg, Sweden.

[20] Mattisson, T., Leion, H., and Lyngfelt, A., (2009), Chemical-looping with oxygen uncoupling using CuO/ZrO2 with petroleum coke, Fuel, 88(4), pp. 683-690.

[21] Ekström, C., Schwendig, F., Biede, O., Franco, F., Haupt, G., de Koeijer, G., . . . Røkke, P.E., (2009), Techno-Economic Evaluations and Benchmarking of Pre-combustion CO2 Capture and Oxy-fuel Processes Developed in the European ENCAP Project, Energy Procedia, 1, pp. 4233-4240.

[22] Anon., *The Costs of CO2 capture – Post-demonstration CCS in the EU – Zero Emission Platform*. 2011, The European Technology Platform for Zero Emission Fossil Fuel Power Plants

[23] Markström, P., Lyngfelt, A., and Linderholm, C., *Chemical-Looping Combustion in a 100 kW unit for Solid Fuels*, in *21st International Conference on Fluidized Bed Combustion*. 2012: Naples.

[24] Markström, P., Linderholm, C., and Lyngfelt, A., *Operation of a 100 kW chemical-looping combustor with Mexican petroleum coke and Cerrejón coal*, in *2nd International Conference on Chemical Looping*. 2012: Darmstadt.

[25] Markström, P., Linderholm, C., and Lyngfelt, A., (2013), Chemical-looping combustion of solid fuels - Design and operation of a 100 kW unit with bituminous coal, International Journal of Greenhouse Gas Control, 15, pp. 150-162.

[26] Markström, P., Linderholm, C., and Lyngfelt, A., (2013), Analytical model of gas conversion in a 100 kW chemical-looping combustor for solid fuels - comparison with operational results, Chem Eng Sci, 96, pp. 131-141.

[27] Linderholm, C., Adanez, J., Beal, C., Epple, B., Penthor, S., Larring, Y., and Lyngfelt, A. (2014) *Chemical-Looping Coal Combustion – Results from the ACCLAIM project*. in *3rd International Conference on Chemical Looping* Göteborg.

[28] Linderholm, C., Knutsson, P., Schmitz, M., Markström, P., and Lyngfelt, A., (2014), Material balances of carbon, sulfur, nitrogen and ilmenite in a 100 kW CLC reactor system, International Journal of Greenhouse Gas Control 27, pp. 1312-1324.

[29] Linderholm, C., Schmitz, M., Knutsson, P., Källén, M., and Lyngfelt, A., (2014), Use of low-volatile fuels in a 100 kW chemical-looping combustor, accepted for publication in *Energy & Fuels*.

[30] Lyngfelt, A. and Leckner, B. (2014) *A 1000 MW_{th} Chemical-Looping Combustor for solid fuels – discussion of design and costs*. in *3rd International Conference on Chemical Looping*. Göteborg, Sweden.

[31] Markström, P. and Lyngfelt, A., (2012), Designing and operating a cold-flow model of a 100 kW chemical-looping combustor, Powder Technology, 222, pp. 182-192.

[32] Kolbitsch, P., Pröll, T., and Hofbauer, H., (2009), Modeling of a 120 kW chemical looping combustion reactor system using a Ni-based oxygen carrier, Chemical Engineering Science, 64(1), pp. 99-108.

[33] Abad, A., Adánez, J., de Diego, L., Gayán, P., García-Labiano, F., and Lyngfelt, A., (2013), Fuel reactor model validation: assessment of the key parameters affecting the chemical-looping coal combustion process, Int. J Greenhouse Gas Control, 19, pp. 451-551.

[34] Arjmand, M., Leion, H., Mattisson, T., and Lyngfelt, A., (2014), Evaluation of different manganese ores in chemical-looping combustion (CLC) for solid fuels, Applied Energy, 113, pp. 1883–1894.

[35] Frohn, P., Arjmand, M., Azimi, G., Leion, H., Mattisson, T., and Lyngfelt, A., (2013), On the High Rate of Gasification of Brazilian Manganese Ore as Oxygen Carrier in Chemical-Looping Combustion (CLC) for Solid Fuels, AIChE J., 59, pp. 4346-4354.

[36] Kolbitsch, P., Bolhàr-Nordenkampf, J., Pröll, T., and Hofbauer, H., (2010), Operating experience with chemical looping combustion in a 120 kW dual circulating fluidized bed (DCFB) unit, International Journal of Greenhouse Gas Control, 4(2), pp. 180-185.

[37] Lyngfelt, A., Pallarés, D., Linderholm, C., Rydén, M., Mattisson, T., , *"Fördelare av gaser i fluidiserad bädd" ("Distributor of volatile gasis in the bottom part of a fluidized bed"), Swedish patent application No. 1400085-5*. 2014.

[38] Yue, G., Lu, J., Zhang, H., Yang, H., Zhang, J., Liu, Q., . . . Jaud, P., *Design theory of circulating fluized bed boilers*, in *18th International Conference on Fluidized Bed Combustion*. 2005: Toronto, Canada.

[39] Haibo, W., Man, Z., Yunkai, S., Qinggang, L., and Yongjie, N., (2012), Research on the heat transfer model of platen heating surface of 300 MW circulating fluidized bed boilers, Powder Technology, 226 pp. 83–90.

[40] Johnsson, F. and Leckner, B., *Vertical distribution of solids in a CFB-furnace*, in *13th Int. Conf. Fluid. Bed Comb.* 1995: Orlando. p. 671-679.