Chemical-Looping Combustion of Solid Fuels – What is Needed to Reach Full-Scale ?

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Abstract – Because the CO_2 capture is inherent in chemical-looping combustion (CLC), thus ideally avoiding costly gas separation, this process has a potential for uniquely low costs of CO_2 capture. So what is needed to get to the realization of this technology? The purpose of the paper is to discuss the status of the technology, barriers to the implementation of the technology, and also to suggest routes for the critical path from successful testing in small pilots to implementation in commercial-sized units.

Thus, operational experiences with oxygen carriers and chemical-looping with solid fuels are discussed, as well as large scale design and important technology challenges. Moreover, possible routes to scale-up are suggested. One way of lowering the costs of intermediate scale-up steps is to build CLC plants without CO_2 purification/compression and oxygen production, because CO_2 capture normally only makes sense in large scale. Another way to avoid or minimize the cost of the air reactor, would be by using a CFBB (circulating fluidized bed boiler) as the air reactor. This could either be an existing CFBB which is not in operation or can be taken out of operation for a period, or a designed dual purpose air reactor/CFBB where the CFBB can be used as a stand-alone unit after the testing period with CLC.

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. Ideally, no gas separation equipment is needed and no energy is expended for gas separation. The CLC system is normally composed of two interconnected fluidized bed reactors, an air and a fuel reactor. Oxygen carriers in the form of metal oxide particles transfer oxygen between the two reactors. Thus, the fuel reacts with the oxygen carrier in the fuel reactor, and forms CO_2 and

H_2O, and after condensation of the H_2O the flow coming from the fuel reactor will consist of more or less pure CO_2. The oxygen carrier is then regenerated in the air reactor.

With solid fuels, the reaction between the char remaining after the release of the volatiles and the oxygen carrier is not direct but involves an intermediate gasification step, as outlined in Figure 1.

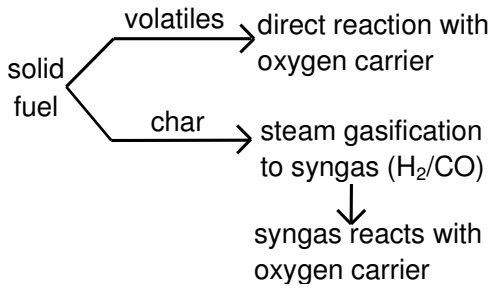

Figure 1. Solid fuel reactions in CLC

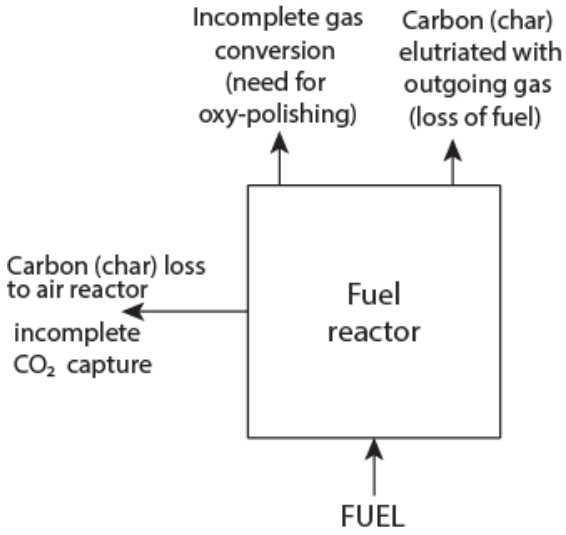

Figure 2. Inadequate performance of the fuel reactor.

Both release of volatiles and char gasification have important implications for the design of the fuel reactor. Ideally, the fuel will be completely converted to CO_2 and H_2O in the fuel reactor. In the case of solid fuels, a fully oxidized gas is normally not attained, which can be remedied by adding oxygen in a post-oxidation chamber downstream of the fuel reactor, so-called "oxy-polishing". Figure 2 illustrates the three deviations from the ideal case; *i)* loss of combustible gases like H_2, CO and CH_4 in the gas leaving the fuel reactor; *ii)* loss of char to the air reactor; and *iii)* loss of char with the gas leaving the fuel reactor. The corresponding performance indicators are:

i. Oxygen demand, Ω_{OD}, is the oxygen required to oxidize unburnt gas leaving the fuel reactor to CO_2 and H_2O over the total oxygen needed to oxidize the gases released from the fuel in the fuel reactor. $1-\Omega_{OD}$ is the gas conversion. The oxygen demand indicates the oxygen needed for oxy-polishing.
ii. CO_2 capture, η_{CO2}, is the fraction of gaseous carbon leaving the fuel reactor related to the total carbon converted to gas in fuel and air reactors. Thus, $1-\eta_{CO2}$ represents the ratio of the carbon lost as CO_2 from the air reactor to the total carbon in gaseous compounds leaving air and fuel reactors.
iii. Fuel conversion, η_F, is used to indicate the char conversion and is defined as the ratio of carbon converted to gaseous compounds in the fuel and air reactors to total carbon added. Consequently, $1-\eta_F$ is the fraction of total carbon added that is elutriated from the fuel reactor in the form of char.

1.1 General barriers for chemical-looping technology:

There are a number of general barriers for implementing CLC technology:

1) At present, the climate targets decided at the Paris meeting in 2015, have not yet been translated into adequate incentives to accomplish the needed reduction in emissions.
2) The process is not well known and understood outside the small CLC community. The CLC concept is a fundamentally novel principle of converting fuels. Although a full-scale CLC boiler could have a design fairly similar to a circulating fluidized bed (CFB) boiler, the reactions happening inside are not normal combustion. The knowledge, understanding and experience of this process, however, mainly resides with a few research groups that have been operating CLC pilots.
3) A related barrier is the risk of investment in a new and largely unproven technology. Closely related to this are the inherent difficulties in the scaling-up of fluidized-bed technologies. It is evident that the results in small pilots are not directly translatable to the large scale. Thus, extrapolation of performance in the small scale to large scale is uncertain. This also means that there are uncertainties regarding the adequate full-scale design. Previous work with a 1000 MW_{th} design shows one possibility, [1], but a fully optimized design could look different.
4) Development of a technology like CLC would be easiest if it could proceed in small steps, going successively from smaller to larger sizes, and build on accumulated experiences. However, in order to meet climate goals, CO_2 capture technology is needed soon and moreover CO_2 capture normally only makes sense in a large scale, and in direct connection with the possibilities to store the CO_2.

Below are a few reflections on these barriers:

1) If there should be any reasonable chance of meeting the climate targets, strong incentives will be necessary in a fairly short time. Hopefully, the Paris agreement will be followed by the action needed to fulfil the agreement.
2) It is important that the CLC community communicates the good results and the applicability of the results and develops a close cooperation with the industry and society.
3) Here it is necessary that mechanisms for financing this development are in place, which will be dependent on adequate incentives in combination with relevant measures to support the phase of up-scaling.
4) The need to have an accelerated development with a reduced number of steps to reach full scale, is facilitated by the large experience with the closely related CFB technology. Further, there could be opportunities to facilitate the demonstrations as will be discussed in section 6.

2 Low-cost oxygen carriers – operating experience

Today, chemical-looping operation in the literature involves at least 8400 h in more than 25 pilots. This only covers operation with fuel. A majority of the operational experiences with chemical-looping combustors uses gaseous fuels and manufactured oxygen carriers. The operational experience in pilots with manufactured materials is more than 6400 h, of which materials based on Ni has been mostly used, cf. Table 1. A number of previous literature reviews have presented the advantages and disadvantages with the various monometallic oxides, e.g. [2], [3, 4]. In recent years there has been an increased interest in combined oxides, usually based on manganese combined with e.g. Ca, Fe, Si.

In the case of chemical-looping operation with solid fuels low-cost materials like ores or waste materials are often used. The reason for this difference is firstly that solid fuels normally contain significant quantities of ash which is expected to lower the possible life-time of the oxygen carrier, and secondly that the hydrocarbons of typical gaseous fuels like natural gas require more reactive oxygen carrier materials than the gas released from solid fuels, which contains a lot of the more reactive gases H_2 and CO. Low-cost materials include iron ores and iron-based waste materials, ilmenite, and manganese ores.

Table 1. Reported/estimated operational experiences from 150 publications.

Manufactured materials:	Reported operational time at least
Nickel	3067
Copper	822
Manganese	91
Iron	1652
Cobalt	178
Combined oxides	646
Ores or waste materials:	
Iron	775
Ilmenite	945
Manganese	183
Calcium sulfate	75

Below an overview of the work reported with low cost materials will be given.

Reported use of iron ores or iron-based waste materials are given in Table 2. The obvious advantage of iron materials is the low cost and the large availability. A number of different iron ores have been successfully used in operation, as well as some iron-based waste materials. As can be seen from the table below most of the operation reported is from smaller units in the 1 kW scale. Some studies have also used additives to improve reactivity. A majority of the work has been done at CSIC in Spain and in Nanjing. Interestingly, the work also includes results from a 50 kW pressurized unit.

Table 2. Operation with Fe-based low-cost materials

Place	Unit	Oxygen carrier	Time, h	Fuel	Ref.
Chalmers	0.3 kW	Fe_2O_3 shells	37	SG	[5]
CSIC	0.5 kW	Fe_2O_3 waste mtrl	111	CH_4, SG	[6]
CSIC	0.5 kW	iron ore (S)	50	CH_4, SG, PSA off-gas	[7]
Nanjing	1 kW – SF	iron ore (Au)	10	coal	[8]
Nanjing	1 kW – SF	iron ore (Au)	?	sawdust/coal	[9]
Nanjing	1 kW – SF	iron ore (Au) + 3% of $NiO/NiAl_2O_4/Al_2O_3$ 20/39/41	10	coal	[10]
Nanjing	1 kW – SF	iron ore iron ore w 4.5% NiO, Imp iron ore w, 6.7% NiO, Imp iron ore + 4.5% NiO iron ore + 6.7% NiO	68	coals	[11]
Nanjing	1 kW – SF	iron ore	20	coal/anthracite	[12]
Nanjing	1 kW – SF	iron ore + K	5	coal	[13]
Nanjing	1 kW – SF	iron ore	22	coal with high K	[14]
Nanjing	1 kW – SF	iron ore	10	sewage sludge	[15]
Nanjing	1 kW – SF	iron ore + cement/CaO	15	coal	[16], [17]
CSIC	0.5/1.5 kW-SF	Fe waste mtrl	40	coal	[18]
CSIC	0.5/1.5 kW-SF	Fe ore (S)	78	biomass	[19]
CSIC	0.5/1.5 kW-SF	Fe ore (S)	30	coals	[20]
CSIC	0.5/1.5 kW-SF	Fe ore (S)	18	anthracite, lignite	[21]
Chalmers	100 kW-SF	Fe ore (S)	26	wood char, 2 coals	[22]
Huazhong	1/5 kW –G/SF	Fe ore	100	CH_4	[23]
Huazhong	1/5 kW – G/SF	Fe ore	100	coal	[24]
Nanjing	25 kW-SF	Fe Ore	>6	rice husk	[25]
Nanjing	50 kW-Pr SF	Fe Ore	19	coal	[26]

SF = solid fuel, GSF=gas/solid fuel, Pr=pressurized

Table 3. Operation with ilmenite

Place	Unit	Oxygen carrier	Time, h	Fuel	Ref.
Chalmers	0.3 kW	ilmenite (N) w Ni	83	natural gas	[27]
Chalmers	0.3 kW	ilmenite (N)	85	syngas	[5]
Chalmers	10 kW–SF	ilmenite (N)	22	coal	[28]
Chalmers	10 kW–SF	ilmenite (N)	11	petcoke	[29]
Chalmers	10 kW–SF	ilmenite (N)	18	petcoke	[30]
Chalmers	10 kW–SF	ilmenite (N)	26	petcoke	[31]
Chalmers	10 kW–SF	ilmenite (N)	4	petcoke	[32]
Chalmers	10 kW–SF	ilmenite (N) + lime	4	petcoke	[32]
Chalmers	10 kW–SF	ilmenite (N)	29	petcoke, coal	[33]
Vienna UT	140 kW	ilmenite (N)	160	CH_4, CO, H_2	[34], [35], [36]
Stuttgart	10 kW	ilmenite (Au)	?	syngas	[37]
CSIC	0.5/1.5 kW-SF	ilmenite (N)	26	coal	[38]
CSIC	0.5/1.5 kW-SF	ilmenite (N)	35	coal	[39]
CSIC	0.5/1.5 kW-SF	ilmenite (N)	30	coal	[40]
CSIC	0.5/1.5 kW-SF	ilmenite (N)	44	coals	[41]
CSIC	0.5/1.5 kW-SF	ilmenite (N)	35	lignite	[42]
Chalmers	0.3 kW LF	ilmenite (N)	80	kerosene	[43]
Chalmers	100 kW-SF	ilmenite (N)	24	coal, PC	[44] [45] [46] [47]
Chalmers	100 kW-SF	ilmenite (N)	12	wood char	[48]
Chalmers	100 kW-SF	ilmenite (N)	34	wood char, PC	[49]
Chalmers	100 kW-SF	ilmenite (N) + Mn ore	18	wood char, 2 PC	[50]
Hamburg	25 kW –SF	ilmenite (Au)	>60	CH_4, coal (21 h)	[51]
Chalmers	10 kW LF	Ilmenite (N)	66	fuel oil	[52]
CSIC	50 kW-SF	Ilmenite (N)	4	coal	[53]
CSIC	50 kW-SF	Ilmenite (N)	30	coal	[54]
Darmstadt	1 MW	Ilmenite (N)	2	coal	[55]
Darmstadt	1 MW	Ilmenite (N)	3	coal	[56]

SF = solid fuel, LF=liquid fuel, NG=natural gas, PC=pulverized coal

Ilmenite is an iron-titanium oxide mineral, $FeTiO_3$, and the term is also used for the natural ores which contain this mineral. Ilmenite is the reduced form of the oxygen carrier and will therefore be oxidized when introduced in a chemical-looping combustor system.

The total operating time with ilmenite is somewhat larger than with iron materials. Here, all of the work has been done in Europe, and with a few exceptions the same Norwegian ilmenite has been used, Table 3. As compared to iron materials, more of the ilmenite operation has been done in larger pilots.

The operational experience with manganese ores is much smaller than with ilmenite or iron ores, see Table 4. This is a bit surprising as manganese ores generally appear to be more reactive than both iron ores and ilmenite. A possible explanation could be that the manganese ores are less easy to access. Most of the work has been done at Chalmers.

Table 4. Operation with manganese -based low cost materials

Place	Unit	Oxygen carrier	Time, h	Fuel	Ref.
Chalmers	0.3 kW	Mn ore	2	NG	[57]
Chalmers	10 kW–SF	Mn ore	10	petcoke	[33]
Chalmers	10 kW–SF	Mn ore + lime	15	petcoke	[58]
Chalmers	10 kW–SF	3 Mn ores	42	wood char, pet coke	[59]
Chalmers	100 kW-SF	Mn ore	52	coal, wood char,	[60]
IFP-Lyon	10 kW-GSF	Mn ore	90	coal, CH_4, syngas	[61]
Tsinghua	1 kW	Mn ore, Mn ore + Cu	182	CO	[62]

SF = solid fuel, GSF=gas/solid fuel

Published results with calcium sulphate as oxygen carrier are only available from Alstom's 3 MW pilot, see Table 5. A challenge with using calcium sulphate is controlling release of sulphur.

Table 5. Other low cost materials

Place	Unit	Oxygen carrier	Time, h	Fuel	Ref.
Alstom	3 MW-SF	$CaSO_4$	75	coal	[63]

2.1 Comparisons

The literature presented above indicates that a number of low-cost materials have been shown to work in actual operation. As the reported work has been done in a number of different units at different conditions, general comparisons are not so easy. However, a comparison of two iron materials and ilmenite has been done at CSIC, indicating better performance of the two iron materials. On the other hand, Chalmers has compared one of these two iron materials, an iron ore, with ilmenite and found that it was possible to reach higher conversion with the iron ore, but only when circulation was significantly increased. It is likely not relevant to indicate any winner in the comparison of ilmenite and iron materials.

Moreover, it is possible to compare the results with ilmenite and manganese ores at the two units at Chalmers. Here the results are quite clear, the manganese ores clearly give a higher conversion of the gas, which is also consistent with laboratory investigations, [64]. Moreover, the presence of potassium in manganese ores promotes the steam gasification of the char, [65]. On the other hand manganese ores show higher loss of fines compared to ilmenite, although the attrition rates vary between different manganese ores.

2.2 Operating experiences with solid fuels

Totally, more than 2900 h of operation with solid fuels has been reported, of which 1340 h with low cost oxygen carriers, as reported in the previous section. Consequently, the operation with solid fuels also involves a lot of work with manufactured materials, involving calcium manganites, nickel materials, copper materials, and iron materials, see Table 6. Of particular interest are the results with copper materials, where very high performance, i.e. complete gas and fuel conversion can be reached because these materials have CLOU properties. CLOU, or Chemical-Looping with Oxygen Uncoupling, means that oxygen is spontaneously released when the oxygen carrier is in an environment with low oxygen concentration, as is the case in the fuel reactor, [66]. A drawback with copper materials is the higher cost. Further, the melting temperature of Cu is relatively low, 1089°C, and thus there could be issues with agglomeration at higher temperatures if Cu is formed. Although Cu is not active in the oxygen uncoupling reaction, Cu could be formed by reduction of CuO or Cu_2O with volatiles. Another CLOU material with potentially lower cost is the combined calcium manganite. However, the release of oxygen is considerably smaller and slower than with copper. Operation with calcium manganite showed improved performance compared to low-cost materials, but this material appears not to be suitable for sulphur-containing fuels.

Despite a high reactivity, nickel materials are likely not a good solution with solid fuels because of high cost, toxicity and sensitivity to the presence of sulphur compounds. In contrast to the other pilot work, which has been done in interconnected fluidized beds, the work at Ohio involves a moving-bed fuel reactor. This work also involves a significant fraction of the operational hours in Table 6.

Table 6. Operation with solid fuels and manufactured oxygen carriers.

Chalmers	10 kW–SF	$CaMn_{0.9}Mg_{0.1}O_3$, SD	74	wood char, petcoke	[67], [68]
Nanjing	10 kW –SF	$NiO/NiAl_2O_4$ 33/67, impr	30	coal	[69]
Nanjing	10 kW –SF	NiO/Al_2O_3 35/65, CoPr	100	coal	[70]
Nanjing	10 kW –SF	Fe_2O_3	30	biomass	[71]
Nanjing	10 kW –SF	NiO/Al_2O_3 35/65	100	sawdust	[72]
Nanjing	1 kW – SF	NiO/Al_2O_3 35/65, CoPr	30	coal	[73]
Nanjing	1 kW – SF	$NiO/NiAl_2O_4/Al_2O_3$ 20/39/41, CP	?	coal	[74]
CSIC	0.5/1.5 kW-SF	$CuO/MgAl_2O_4$ 60/40, SD	40	coal	[75]
CSIC	0.5/1.5 kW-SF	$CuO/MgAl_2O_4$ 60/40, SD	40	coals	[76]
CSIC	0.5/1.5 kW-SF	$CuO/MgAl_2O_4$ 60/40, SD	15	lignite	[77]
CSIC	0.5/1.5 kW-SF	$CuO/MgAl_2O_4$ 60/40, SD	10	biomass	[78]
Ohio	2.5 kW-SF	supported Fe_2O_3	>300	coal	[79], [80], [81], [82], [83], [84]
Ohio	25 kW –SF		>300 SLC	CH4, syngas	
Ohio	25 kW –SF		>230 CDLC	coal	

3 Performance in larger pilots

Table 7 shows a comparison of operational performance in the four largest pilots. Because of the lack of detail in the data from the 3 MW unit, the focus will be on the other three. The performance of the 1 MW unit was studied using ilmenite and two quite different sizes of coal. The first case was pulverized coal (PC) and the second was larger coal (LC) particles of up to 8 mm size, and in both cases a large loss of carbon from the fuel reactor was observed, either to the air reactor or elutriated:

- For pulverized coal there is a very large loss of elutriated carbon from fuel reactor, 50%, in addition to the loss to the air reactor, indicated by a carbon capture of 80%.
- For the larger coal there is instead a very large loss of char to the air reactor, indicated by a carbon capture of only 50%. Thus, residence time is too short to convert the larger coal particles in the fuel reactor.

Operation of pulverized coal and ilmenite was also investigated in the 100 kW unit. The loss of carbon by elutriation from the 100 kW unit was also high, but lower than in the 1 MW unit, around 35%. Further, the loss of carbon to the air reactor was low, as indicated by the high carbon capture. Moreover, the oxygen demand is somewhat lower in the 100 kW unit. Results for the 100 kW with a mixture of ilmenite and a manganese ore showed similar results for carbon capture and loss of carbon by elutriation. However, lower oxygen demand was attained.

Operation using an intermediately sized coal (IC) in the 100 kW unit showed a significantly lowered loss of carbon by elutriation, but without any measurable increase in the loss to the air reactor. Further, the oxygen demand was improved, which can be attributed to the use of a more reactive oxygen carrier, a manganese ore.

Finally, results from the 50 kW pilot, operated at 13.5 kW with another coal of intermediate size, also showed low loss from elutriation. Further, the results from the 50 kW unit showed lower oxygen demand but higher loss of carbon to air reactor in comparison to the 100 kW unit.

Table 7. Comparison of larger CLC pilots.

	3 MW Alstom	1 MW Darmstadt, ilmenite		100 kW at Chalmers			50 kW, CSIC
	coal $CaSO_4$ [63]	PC [55][1]	LC [56]	PC, ilm [48]	PC, ilm + Mn ore 1 [50]	IC, Mn ore 2 [60]	IC, ilm, test 6 [54]
Carbon capture, η_{CO2}, [%]	96	80	44-52	98-99	99	99	90
Carbon loss by elutriation, 1-η_F [%]	0.5	50	5	35	(26-46)	**8-12**	**7**
Oxygen demand, Ω_{OD}, [%]		20[1] (26-38)	22-28	17-25	**8,5**-18	11-17	10
Pressure drop fuel reactor, kPa			7.5	14-25			9
Solids inventory, kg/MW		156	105	300-500[2]			480
T FR, ºC		900	920-950	965-980	960-974	970-980	990

[1] Not isothermal. Propane and air added to fuel reactor to keep up temperature. [2]fuel reactor, PC = pulverized coal: a majority below 90 μm, LC = larger coal, <8 mm, IC = intermediate sized coal: a majority in the size range 90-300 μm.

The differences between the results at Chalmers and in Darmstadt appear to be possible to explain as follows:

- The circulation in the 1 MW unit is not sufficient to reach adequate fuel reactor temperature, resulting in slow char conversion. This increases loss of char.
- The solids inventory in the fuel reactor of the Darmstadt unit is low, resulting in short residence time for char to convert, i.e. larger loss to air reactor. It also means less contact between reacting gas and oxygen carrier, resulting in higher oxygen demand.
- The autothermal testing with "large coal" was done without carbon stripper in order to reduce cooling. Thus, it was possible to attain auto-thermal operation. This also contributes to high loss to AR, but the main reason is likely the fuel size. The PC firing on the other hand was made with a carbon stripper, nevertheless there was a significant loss to the air reactor.
- The fuel inlet in the 1 MW is high, 0.69 m above the nozzle plate, meaning that fuel is fed in the uppermost part of the dense bottom bed. This gives poorer contact between volatiles released and oxygen carrier, i.e. higher oxygen demand.

The oxygen demand in the 1 MW is higher than in Chalmers 100 kW, which is expected in view of lower solids inventory, lower temperature and the high fuel entry mentioned above. The much higher loss of char to the air reactor and by elutriation also means that less syngas is formed, which should also raise the oxygen demand. This is because the oxygen carrier is more reactive towards syngas than the hydrogen-containing volatiles, in addition to the volatiles having poorer contact with the bed material. Adding all these effects it is reasonable to expect a larger difference in oxygen demand than what was observed. However, the fluidizing conditions in the Darmstadt unit are significantly better with a riser twice as high with a significant pressure drop, where good contact between gas and solids is expected. This is in contrast with the low ratio of diameter / bed height in the 100 kW unit giving slugging conditions. In conclusion it would be expected that raised solids inventory, feeding of fuel in the low part of the bed in combination with higher temperature would give a significant improvement and most likely lower oxygen demand as compared to the 100 kW unit.

The following can be concluded from the comparison:

- The fuel size is critical, pulverized coal gives large losses of unconverted char, whereas large fuel particles results in poor CO_2 capture. However, with an intermediate size it possible to achieve high char conversion in the fuel reactor as indicated by the results in the 50 and 100 kW pilots. Thus a size range of around 100 to 300 μm is probably optimal.
- To reach good CO_2 capture, sufficient temperature and solids inventory are needed in the fuel reactor.
- The fuel should be fed as low as possible in the fuel reactor bed to achieve good contact between bed material and volatiles released.
- The results from the two smaller units indicate that good or reasonable performance could be reached simultaneously for all the three key performance criteria. This was not the case for the 1 MW unit. However, the reasons for inadequate performance are well understood as well as the measures needed to improve performance.

4 Proposed designs for scale-up

Presently, proposed designs of solid-fuel chemical-looping boilers presented in the literature involve a 100 MW_{th} and a 1000 MW_{th} unit, see Fig. 3a and b, [85] [1]. The design of the smaller unit is not linked to any existing CFB units but based on more fundamental modeling, whereas the design of the 1000 MW_{th} unit was done with the purpose of showing that a design can be made similar to large CFB boilers. Thus, this unit has essentially similar gas velocities, cross-sections, number of cyclones etc. as the 460 MW_e Lagisza plant. The important difference compared to the Lagisza plant is the splitting of the cross-section in three parts to give a centrally placed fuel reactor, see Fig. 3b. Further it was motivated to lower the heights of the air reactors. The similarities of the CLC and CFB plants also facilitated the estimation of the added costs of the CLC as compared to a CFB. Thus, the cost was estimated to around 20 €/tonne CO_2, where oxygen production and CO_2 purification/compression are the main costs.

In contrast to the 100 MW_{th} design, the 1000 MW_{th} design does not include any carbon stripper. Also the principle for returning the circulating material from fuel reactor to air reactor are different. Otherwise, the general principle of units is similar. The smaller unit has lower riser heights and despite the smaller size more, *i.e.,* 8 cyclones. Moreover, velocities are somewhat lower in the smaller unit giving a higher cross-sectional area per MW. The added gas flow from the carbon stripper further increases the cross-section of the fuel reactor.

In case of the 1000 MW_{th} design it can be noted that there is a symmetry over the cross-section in both directions. Thus, this design should be possible to downscale by a factor of four, i.e. 250 MW_{th}, as indicated by the dashed line in Fig. 3b.

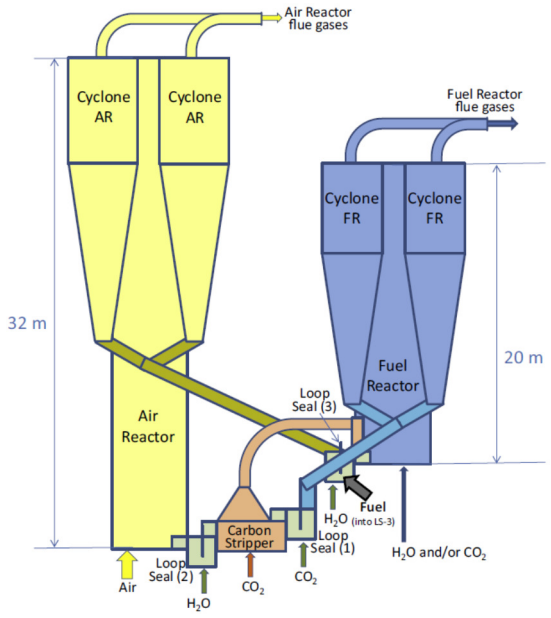

Fig. 3a. 100 MW_{th} chemical-looping boiler design.

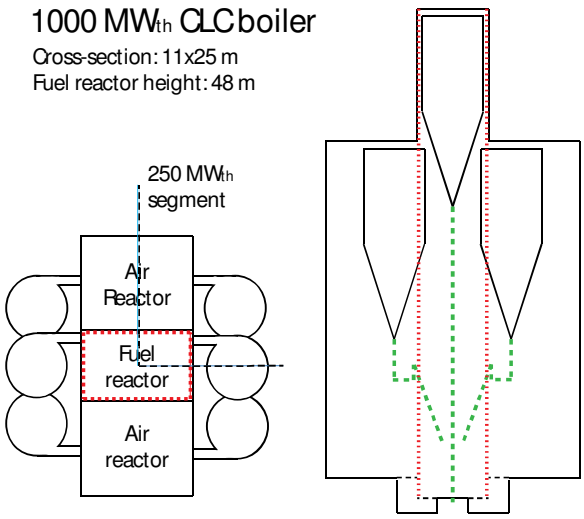

Fig. 3b. 1000 MW_{th} chemical-looping boiler design.

In conclusion, designs have been published showing how full-scale CLC systems can be dimensioned with consideration of mass balances, heat balances and fuel conversion.

5 Important technology challenges

Clearly, the experiences and know-how from CFB technology are central for CLC. However, there are also a number of important differences that have to be properly addressed to make the CLC technology work:

1) Adequate circulation and control of circulation. For the operation of a CLC unit, adequate circulation is a necessity. There are several ways to design and operate an air reactor to control the circulation as discussed by Lyngfelt & Leckner, [1]. This includes fluidizing velocity, reactor height, oxygen carrier size. Another possibility is a separate, dedicated riser.
2) Suitable oxygen carrier materials. The literature cited above shows that a number of low cost materials have been shown to work well in pilot operation.
3) Gas conversion in fuel reactor. Scale-up of the fuel reactor, assuming an internally circulating fluidized bed, will mean a higher riser. Increased height involves increased pressure drop in the riser, i.e. the gas will be in contact with more oxygen carrier, which should improve conversion.
4) Loss of char to air reactor and by elutriation. As discussed above adequate fuel size, sufficient solids inventory and sufficient temperature are needed.
5) Downstream treatment. After the fuel reactor an oxygen polishing step is foreseen, where the gas is fully oxidized to H_2O and CO_2, and also reduced sulphur and nitrogen compounds are oxidized. This step will produce a gas similar to that of oxy-combustion, albeit with considerably less argon. Thus, the gas from the oxy-polishing step can be treated in a similar way as planned for oxy-fuel process. This involves downstream steps for making the CO_2 stream pure enough for storage, which would involve condensation and removal of moisture, removal of acid gases and oxygen, and CO_2 compression.

6 Routes for scale-up at reduced costs

The scaling-up of CLC to commercial size will need intermediate steps, which should verify the performance of the CLC process under conditions relevant for larger scale. In these steps CO_2 capture would normally have little relevance. This is an advantage and a simplification, avoiding process steps which are not necessary without CO_2 capture, like CO_2 purification and compression and oxygen production. The latter is not needed if the fuel reactor gas is oxidized with air.

Nevertheless, even without these process steps, the costs of building a dedicated CLC demo of intermediate size would be significant. However, there could be options to further reduce demonstration costs:

- Adaptation of an existing CFB. Here the idea is to find an existing CFB unit of suitable size and design which is not in use or can be taken out of operation for a period. This would then be equipped with a fuel reactor.
- Dual purpose CLC/CFB. Here the idea is to build a CLC reactor system to demonstrate the CLC technology, but where the air reactor with some modifications can be used as a CFB boiler after the demonstration period.

The strategy in these approaches is to avoid or minimize the costs of air reactor and the peripheral systems. The fuel reactor is necessary in CLC and no use except in CLC is likely, which means that the cost of the fuel reactor is not easily avoided. The fuel reactor itself is expected to have moderate costs because the walls are adiabatic. For example cost of the fuel reactor walls of a 1000 MW_{th} unit including cyclones, channels etc., was estimated to 4 M€, [1]. This is in fact only a few per cent of the estimated boiler cost.

7 Conclusions

The necessary elements for a scale-up are at hand:

- Different oxygen carrier materials of low cost have been tested in extended operation and found to have reasonable performance with respect to reactivity and life time.
- Operation with solid fuels in pilots up to at least 100 kW has been shown to work. Operation in a 1 MW pilot showed inadequate loss of char but reasons for this are understood and can be addressed.
- Designs for large-scale units have been done indicating that the process is technically realistic and should have low cost of CO_2 capture.
- A scale-up strategy to minimize costs has been suggested.

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