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# 11,000 h of Chemical-Looping Combustion Operation – Where Are We and Where Do We Want to Go?

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## Abstract

A key for chemical-looping combustion (CLC) is the oxygen carrier. The ultimate test is obviously the actual operation, which reveals if it turns to dust, agglomerates or loses its reactivity or oxygen carrier capacity. The CLC process has been operated in 46 smaller chemical-looping combustors, for a total of more than 11,000 h. The operation involves both manufactured oxygen carriers, with 70% of the total time of operation, and less costly materials, i.e. natural ores or waste materials. Among manufactured materials, the most popular materials are based on NiO with 29% of the operational time, Fe<sub>2</sub>O<sub>3</sub> with 16% and CuO with 13%. Among the monometallic oxides there are also Mn<sub>3</sub>O<sub>4</sub> with 1%, and CoO with 2%. The manufactured materials also include a number of combined oxides with 11% of operation, mostly calcium manganites and other combined manganese oxides. Finally, the natural ores and waste materials include ilmenite, FeTiO<sub>3</sub> with 13%, iron ore/waste with 9% and manganese ore with 6%. In the last years a shift towards more focus on CuO, combined oxides and natural ores has been seen.

The operational experience shows a large variation in performance depending on pilot design, operational conditions, solids inventory, oxygen carrier and fuel. However, there is at present no experience of the process at commercial or semi-commercial scale, although oxygen-carrier materials have been successfully used in commercial fluidized-bed boilers for Oxygen-Carrier Aided Combustion (OCAC) during more than 12,000 h of operation.

The paper discusses strategies for upscaling as well as the use of biomass for negative emissions. A key question is how scaling-up will affect the performance, which again will determine the costs for purification of CO<sub>2</sub> through e.g. oxy-polishing. Unfortunately, the conditions in the small-scale pilots do not allow for any safe conclusions with respect to performance in full scale. Nevertheless, the experiences from pilot operation shows that the process works and can be expected to work in the large scale and gives important information, for instance on the usefulness of various oxygen-carriers. Because further research is not likely to improve our understanding of the performance that can be achieved in full scale, there is little sense in waiting with the scale-up.

A major difficulty with the scaling-up of a novel process is in the risk. First-of-its-kind large-scale projects include risks of technical mistakes and unforeseen obstacles, leading to added costs or, in the worst case, failure. One way of addressing these risks is to focus on the heart of the process and build it with maximum flexibility for future use. A concept for maximum flexibility is the Multipurpose Dual Fluidized Bed (MDFB). Another is to find a suitable existing plant, e.g. a dual fluidized-bed thermal gasifier.

With present emissions the global CO<sub>2</sub> budget associated with a maximum temperature of 2°C may be spent in around 20-25 years, whereas the CO<sub>2</sub> budget for 1.5°C is may be exhausted in 10 years. Thus, the need for both CO<sub>2</sub> neutral fuels and negative emissions will become increasingly urgent as we are nearing or transgressing the maximum amount of CO<sub>2</sub> that can be emitted without compromising the global climate agreement in Paris saying we must keep “well below” 2°C and aim for a maximum of 1.5°C. Thus, biomass may turn out to be a key fuel for Carbon Capture and Storage (CCS), because CO<sub>2</sub>-free power does not necessarily need CCS, but negative emissions will definitely need Bio-CCS.

## 1 Introduction

### 1.1 Previous reviews on chemical-looping

The purpose of this paper is not to reiterate previous review papers concerning chemical-looping combustion (CLC), with detailed descriptions of basic concepts. However, for readers not acquainted with these concepts a number of previous reviews can be recommended.

Among the most comprehensive reviews of chemical-looping is a paper by Adánez et al. [1] and a book edited by Fennel and Anthony, [2], containing chapters on fundamentals and reactor design, oxygen carriers, gaseous fuels, liquid fuels, solid fuels and hydrogen production. Additional reviews on chemical-looping combustion can be found in [3],[4], [5],[6],[7].

There are a number of reviews with more focus on oxygen carriers, e.g. [8],[9]. An overview of laboratory examinations, [10], includes 600 oxygen-carrier materials and an update of this study includes another 300 materials, [11]. Material overviews can also be found in PhD theses, e.g. [12-15].

Reviews with focus on chemical-looping combustion of solid fuels are found in, [16], [17], [18], [19], [20], [21].

There are also papers showing the design of existing chemical-looping combustors and the results from operation of these using various oxygen-carrier materials e.g. [22], [1],[23].

Further, there are reviews with more focus on process concepts, [24], including one book on chemical-looping systems [25].

An important aspect of oxygen carriers is the thermodynamic properties, [26], these have also been examined for the more complex combined manganese materials, [27]. These materials have the ability to release oxygen to the gas phase, albeit to varying extent. This variety of the chemical-looping reaction mechanism is commonly referred to as CLOU or Chemical-

Looping with Oxygen Uncoupling, and was first proposed by Mattisson et al., [28], and has later been reviewed [29], [30].

The mechanical integrity is an important property. In two papers with very similar approach, lifetime of oxygen carrier particles derived from actual operation, has been compared to data from attrition testing and crushing testing, [31], [32]. The main conclusion from these papers is the same: Although attrition testing and crushing testing may provide a reasonable idea on which materials to exclude from further testing, the results from these tests are not very well correlated to actual performance. Thus, to show the usefulness of a material there seems to be no other way than prolonged operation in an actual chemical-looping combustor.

This paper will focus on the operational experiences with oxygen carriers and also discuss the future development of CLC technology.

## 1.2 Principal definition and subdivisions of chemical-looping combustion

In this work, chemical-looping or chemical-looping combustion is used solely in connection with processes that transfer oxygen in connection with fuel conversion at high temperatures, which is in accordance with convention. Chemical-looping combustion can be used for both gaseous, liquid and solid fuels, Fig. 1. The definition used here is based on the state of the fuel entering the chemical-looping process. Thus, gas from gasification belongs with Gas-CLC, as long as the gasification step is external to the actual CLC process.

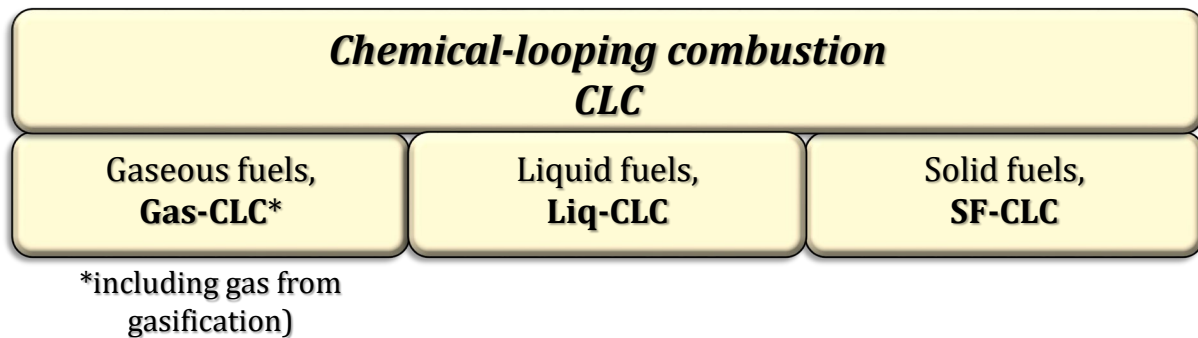


Figure 1. Subdivision of chemical-looping combustion related to fuel.

The reaction mechanism for oxidation of fuel in the fuel reactor could proceed either via heterogeneous reactions, i.e. combustible gases reacting with the oxygen carrier particles, hence called heterogeneous CLC, or by release of oxygen from the oxygen carrier to the gas phase that subsequently reacts with the gaseous, liquid or solid fuel, i.e. Chemical-Looping with Oxygen Uncoupling. Both reaction mechanisms may also happen in parallel, Fig. 2.

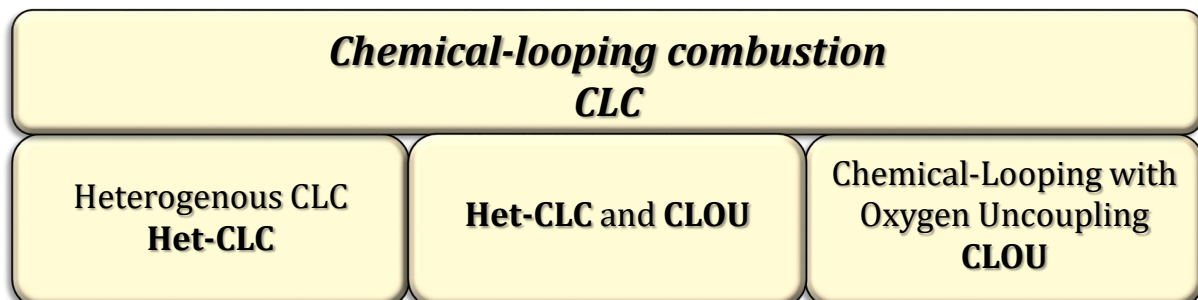


Figure 2. Different reaction mechanisms in the fuel reactor.

Here, CLOU is seen as a possible mechanism of CLC that may improve the conversion of gaseous, liquid and solid fuel. For oxygen carriers with CLOU properties, it is in practice difficult to distinguish between the two mechanisms, so the existence of a pure CLOU mechanism would be difficult to be verified, except for a fuel with absolutely no volatiles. Even if a CLOU material is able to release its oxygen rapidly, can we be sure no heterogeneous reactions take place locally in high concentrations of volatiles or gaseous fuel? Also, the distinction between heterogeneous CLC and mixed CLOU/Het-CLC may not be so clear at times. Thus, a number of materials, normally considered to be non-CLOU materials, have shown minor releases of oxygen. One example is ilmenite, [33].

In conclusion, it is recommended that CLOU is not seen as a different process, but rather as a reaction mechanism that may take place in parallel with heterogeneous CLC. The fraction of each mechanism may be difficult to establish and will be dependent on the fuel, the temperature, the oxygen carrier as well as reactor design and other operational conditions.

Often iG-CLC (in-situ Gasification CLC), is used with the same meaning as SF-CLC, but other times it is used to indicate chemical-looping of solid fuels in a process where no CLOU takes place, i.e. Het-CLC. The name in-situ Gasification CLC could be confusing, as it gives associations to gasification processes, although it is a combustion process where the gasification is not a process step, but rather a mechanism. Further, iG-CLC should not be confused with Integrated Gasification Chemical-Looping Combustion (IGCLC). Therefore, iG-CLC is not used as a term in this paper.

Finally, chemical looping combustion can be divided between different purposes, i.e. energy production and fuel production, Fig. 3. The latter case may involve partial combustion/oxidation, for instance Autothermal Chemical-Looping Reforming, CLR-A, where gaseous or liquid hydrocarbons are partially oxidized to produce a syngas, or Chemical-Looping Gasification, CLG, where solid fuel is converted to combustible gases.

Finally, full oxidation CLC may be combined with conventional steam reforming, by using the off-gas from the reforming process for heating the endothermic steam reforming, CLR-S. CLR-S appears to have significant advantages as compared to CLR-A, [34]. CLR-S is close to conventional reforming, except that heat is transferred from combustion to the reformer tubes using a fluidized-bed heat exchanger instead of gas burners. The more efficient heat transfer makes significantly lower temperature outside of the reforming tubes possible, leading to lower heat losses, i.e. higher reforming efficiency. Whereas the gas from CLR-A eventually needs to be separated in two essentially pure streams, i.e.  $H_2$  and  $CO_2$ , it is sufficient to separate a pure stream of  $H_2$  in CLR-S. The remaining off-gas being burnt in the fuel reactor will, provided gas conversion is complete, yield a pure  $CO_2$  stream after water condensation. CLR-S also makes it possible to have a pressurized steam reforming process, while the chemical-looping process is at ambient pressure. Furthermore, it could be advantageous to have the reforming catalyst contained in reforming tubes, instead of constituting particles in fluidized-beds. This is because reforming catalysts could be less suitable for fluidized-bed operation because of high cost and/or health, safety and environmental concerns.

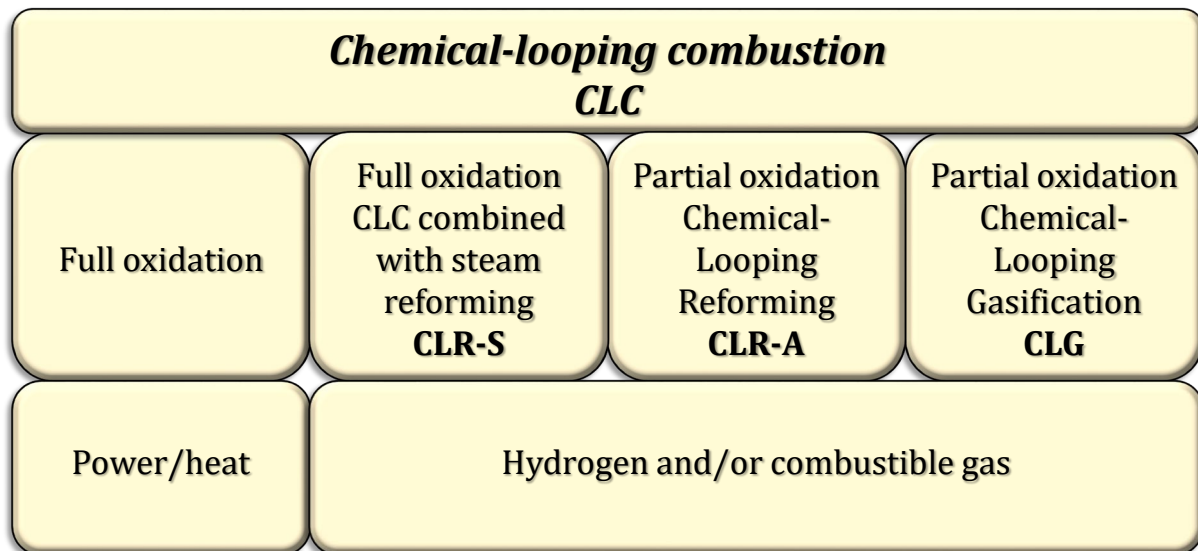


Figure 3. Different applications of chemical-looping.

## 2 Chemical-Looping Operational Experiences

### 2.1 Oxygen carrier materials

#### 2.1.1 Overview of oxygen carrier operation

An overview of operation with oxygen carrier materials as summarized from 213 publications is given in Table 1. A comparison is made to a previous overview where data were collected in the spring of 2014, [9], i.e. four years before the present data. The review from 2014 covers the first ten years of operation of CLC pilot units.

Firstly, it can be noted that the total time of operation is more than 11 000 h. The majority of this operational time is with manufactured materials, i.e. 70%. Manufactured materials are most often used with gaseous and liquid fuels, whereas less than half of the operation with solid fuels uses manufactured materials. For manufactured materials, oxides of Ni dominate the use with gaseous fuels, followed by oxides of copper and combined oxides. For solid fuels the most used materials are manufactured iron oxides, followed by ilmenite and iron ores. Thus, gaseous fuels are associated with the most expensive materials, oxides of Ni and Cu, whereas solid fuels are associated with the cheapest materials, i.e. iron oxides, ores and waste materials, together exceeding 80% of all operation with solid fuels. Among the natural/waste materials the most used oxygen carrier is ilmenite, followed by iron ores/waste and manganese ores.

Table 1: Overview of hours of chemical-looping operation with different oxygen carrier materials.

Type	Oxygen carrier	Gaseous fuel	Liquid fuel	Solid fuel	Total 2018	Total 2018, %	Total 2014	2014-2018	increase '14-'18, %
Manufactured	NiO	2677	377	237	<b>3291</b>	29%	2800	491	18%
	CuO	1130	122	173	<b>1425</b>	13%	627	798	127%
	Mn <sub>3</sub> O <sub>4</sub>	74	17	0	<b>91</b>	1%	91	0	0%
	Fe <sub>2</sub> O <sub>3</sub>	617	77	1072	<b>1766</b>	16%	1077	689	64%
	CoO	178	0	0	<b>178</b>	2%	178	0	0%
	Combined oxides	918	10	289	<b>1217</b>	11%	545	672	123%
Natural ore or waste material	Fe ore	488	0	576	<b>1064</b>	9%	404	660	163%
	Ilmenite	538	150	788	<b>1496</b>	13%	810	686	85%
	Mn ore	354	0	381	<b>735</b>	6%	148	587	397%
	CaSO <sub>4</sub>	0	0	75	<b>75</b>	1%	75	0	0%
Total manufactured		5594	603	1771	<b>7968</b>	70%	5318	2650	50%
Total natural/waste		1380	150	1820	<b>3370</b>	30%	1437	1933	135%
<b>Total</b>		6974	753	3591	<b>11338</b>	100%	6755	4583	68%
<b>Publications</b>					212		115	97	84%

The table also shows the increase during the latest four to five years, i.e. 2014-2018. Here an increase by less than appr. 40-50% would signify a reduced activity as compared to the previous ten years. This is clearly the case with Ni oxides, which dominated the first ten years. In absolute numbers, copper oxides were the most used in the last four-year period, followed by iron oxides, ilmenite, iron ore and combined oxides in fierce competition, and with manganese ore lagging slightly behind. Percentagewise the greatest increase was for manganese ore, followed by iron ore, copper oxide and combined oxides. These four have in common that they are of interest for solid fuels, either being low-cost materials or, in the case of copper oxide, having CLOU properties. Although operation with copper and combined oxides has risen significantly, this does not apply to generally to manufactured materials. This is in contrast to natural/waste materials which have more doubled their operation in the last four years, having a share of 43% of the total operation in this period.

In total, operation time has increased by 68% the last four years and the number of publications on operation has increased by 84%, see Table 1. The yearly number of publications on CLC operation shows a steady increase, see Fig. 4, with bi-annual peaks associated with the CLC and the GHGT (Greenhouse Gas Control Technologies) conferences. The same trend of increase also generally applies to publications on chemical-looping, i.e. publications with “chemical-looping” in title (Scopus), Fig. 4.

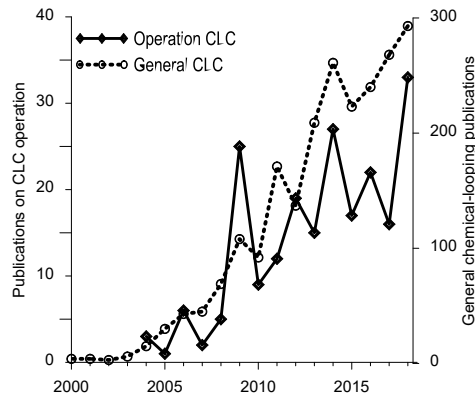


Figure 4. Publications on CLC operation,  $\diamond$ , and “chemical-looping” in title (Scopus),  $\circ$ , vs year published.

An overview of the operation of these oxygen carrier groups will be given in more detail below, starting with Ni-based materials.

### 2.1.2 Ni-based materials

The oxidized and reduced forms are NiO and Ni. Nickel oxide materials were early identified as the oxygen carrier being most reactive with methane, and is also the most studied material. Metallic nickel, the reduced form, is a methane reforming catalyst which likely explains the high reactivity towards methane. This is also supported by laboratory cyclic batch experiments, i.e. in tests where the gradual reduction of NiO by methane can be followed. Initially a quite significant fraction of unconverted methane can be seen, which eventually fall to zero, likely associated with the formation of Ni, [35].

Important disadvantages with nickel materials are HSE (health, safety and environment) concerns and high costs. Moreover, thermodynamics restrict the gas conversion of CH<sub>4</sub> to CO<sub>2</sub> to 99-99.5%, [26]. Table 2 gives an overview of operation with Ni materials.

Table 2: Operation with nickel-based oxides. For details on operators see section 2,2 below.

Operator	Unit	Active oxide/support, prod. method	Time, h	Fuel	Ref.
Chalmers	10 kW	NiO/NiAl <sub>2</sub> O <sub>3</sub> 40/60, FG	100	NG	[36, 37]
Chalmers	10 kW	NiO/NiAl <sub>2</sub> O <sub>4</sub> 60/40, SF	165	NG	[38]
Chalmers	10 kW	NiO/NiAl <sub>2</sub> O <sub>4</sub> 40/60, SD NiO/NiAl <sub>2</sub> O <sub>4</sub> /MgAl <sub>2</sub> O <sub>4</sub> 40/42/18 SD	1000	NG	[39]
KIER	50 kW	NiO/bentonite 60/40, MDCC	3	NG	[40]
Chalmers	0.3 kW	NiO/MgAl <sub>2</sub> O <sub>4</sub> 60/40, FG	30	NG	[41]
Chalmers	0.3 kW	NiO/NiAl <sub>2</sub> O <sub>3</sub> 40/60, FG NiO/MgAl <sub>2</sub> O <sub>4</sub> 60/40, FG	8	NG/SG	[42]
Chalmers	0.3 kW	NiO/MgAl <sub>2</sub> O <sub>4</sub> 60/40, FG	41	NG	[43]
Chalmers	0.3 kW	NiO/MgAl <sub>2</sub> O <sub>4</sub> 20/80, FG	160	NG	[44]
Chalmers	0.3 kW	NiO/ZrO <sub>2</sub> (Mg) 40/60, FG	40	NG	[45]
Chalmers	0.3 kW	NiO/NiAl <sub>2</sub> O <sub>4</sub> 40/60, SD NiO/NiAl <sub>2</sub> O <sub>4</sub> /MgAl <sub>2</sub> O <sub>4</sub> 40/42/18 SD NiO/ $\alpha$ -Al <sub>2</sub> O <sub>3</sub> 18/82, HI	84	NG	[46]
CSIC	0.5 kW	NiO/ $\alpha$ -Al <sub>2</sub> O <sub>3</sub> 18/82, HIWI	70	CH <sub>4</sub>	[47]
CSIC	0.5 kW	NiO/ $\alpha$ -Al <sub>2</sub> O <sub>3</sub> 19/81, HIWI	40	LHC	[48]
CSIC	0.5 kW	NiO/ $\alpha$ -Al <sub>2</sub> O <sub>3</sub> 19/81, HIWI	35	CH <sub>4</sub> + H <sub>2</sub> S	[49]
CSIC	0.5 kW	NiO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 21/79, HIWI NiO/ $\alpha$ -Al <sub>2</sub> O <sub>3</sub> 18/82, HIWI	50	CH <sub>4</sub>	[50] [51]
CSIC	0.5 kW	NiO/ $\alpha$ -Al <sub>2</sub> O <sub>3</sub> 18/82, HIWI	50	SG	[52]
CSIC	0.5 kW	NiO/ $\alpha$ -Al <sub>2</sub> O <sub>3</sub> 19/81, HIWI	35	CH <sub>4</sub>	[53]
CSIC	0.5 kW	NiO/CaAl <sub>2</sub> O <sub>4</sub> 12/88, WI [54]	90	CH <sub>4</sub> , H <sub>2</sub> , CO, SG,	[55]

		NiO/ $\alpha$ -Al <sub>2</sub> O <sub>3</sub> 18/82, HIWI		LHC	
CSIC	0.5/1 kW-LF	NiO/ $\alpha$ -Al <sub>2</sub> O <sub>3</sub> 18/82, HIWI	120	ethanol	[56]
CSIC	0.5/1 kW-LF	NiO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 21/79, HIWI	53	ethanol	[54]
CSIC	0.5/1 kW-LF	NiO/ $\alpha$ -Al <sub>2</sub> O <sub>3</sub> 18/82 NiO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 21/79	50	ethanol	[57]
CSIC	0.5/1 kW-LF	NiO/ $\alpha$ -Al <sub>2</sub> O <sub>3</sub> 18/82	50	Diesel, oil	[58, 59]
CSIC	0.5/1 kW-LF	NiO/ $\alpha$ -Al <sub>2</sub> O <sub>3</sub> 18/82 HIWI NiO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 21/79 IWI	50	bioethanol	[60]
KAIST	1 kW	NiO/bentonite 60/40 NiO/Fe <sub>2</sub> O <sub>3</sub> /bent. 45/15/40 NiO/Fe <sub>2</sub> O <sub>3</sub> /bent. 30/30/40	6	CH <sub>4</sub>	[61]
Vienna UT	140 kW	NiO/NiAl <sub>2</sub> O <sub>4</sub> 40/60, SD NiO/NiAl <sub>2</sub> O <sub>4</sub> /MgAl <sub>2</sub> O <sub>4</sub> 40/42/18 SD NiO/ $\alpha$ -Al <sub>2</sub> O <sub>3</sub> 18/82, HIWI	appr. 240*	NG, CO, H <sub>2</sub>	[62-72]
Vienna UT	140 kW	NiO/NiAl <sub>2</sub> O <sub>4</sub> 40/60, SD + NiO/NiAl <sub>2</sub> O <sub>4</sub> /MgAl <sub>2</sub> O <sub>4</sub> 40/42/18 SD	32	NG +H <sub>2</sub> S	[73]
Alstom	15 kW	NiO/NiAl <sub>2</sub> O <sub>3</sub> 40/60, FG NiO/NiAl <sub>2</sub> O <sub>4</sub> 40/60, SD NiO/NiAl <sub>2</sub> O <sub>4</sub> /MgAl <sub>2</sub> O <sub>4</sub> 40/42/18 SD NiO/ $\alpha$ -Al <sub>2</sub> O <sub>3</sub> 18/82, HIWI	100	NG	[74]
Nanjing	10 kW –SF	NiO/NiAl <sub>2</sub> O <sub>4</sub> 33/67, impr	30	coal	[75]
Nanjing	10 kW –SF	NiO/Al <sub>2</sub> O <sub>3</sub> 35/65, CP	100	coal	[76]
Nanjing	10 kW –SF	NiO/Al <sub>2</sub> O <sub>3</sub> 35/65	100	sawdust	[77]
KIER	50 kW new	NiO/?? 70/30, SD	100-200	NG, SG	[78]
Nanjing	1 kW – SF	NiO/Al <sub>2</sub> O <sub>3</sub> 35/65, CP	30	coal	[79]
Nanjing	1 kW – SF	NiO/NiAl <sub>2</sub> O <sub>4</sub> /Al <sub>2</sub> O <sub>3</sub> 20/39/41, CP	?	coal	[80]
IFP-Lyon	10 kW-GSF	NiO/NiAl <sub>2</sub> O <sub>4</sub> 60/40, Pr	18	CH <sub>4</sub>	[81]
Chalmers	0.3 kW LF	NiO/ZrO <sub>2</sub> (Mg) 40/60, SD	54	kerosene	[82]
Nanjing	25 kW-SF	NiO/Al <sub>2</sub> O <sub>3</sub> 60/40 + sand	>7	rice straw	[83]
KIER	200 kW	70% NiO SD	100	NG, SG	[84]

\*Personal communication from Tobias Pröll.

Abbreviations used in Tables 2-11:

*CLC units:* SF solid fuel, LF liquid fuel, GSF gaseous, liquid and solid fuel; *Materials:* w with (added to particle), + physically mixed.; letters within parenthesis indicate origin, e.g. country or company. *Production methods:* FG freeze granulated, SD spray-dried, HIWI=hot incipient wet impregnation; IWI= incipient wet impregnation; WI=wet impregnation; impr=impregnation; CP=coprecipitation; Pr=precipitation; WM=waste material. *Fuels:* NG natural gas, SG syngas, PC petroleum coke, LHC lower hydrocarbons, e.g. ethane, propane, PSA-OG pressure swing adsorption off-gas.

### 2.1.3 3.3 Cu-based materials

The oxidised form is CuO and the fully reduced form is Cu. Moreover, copper can be used as a CLOU material, with the reduced form Cu<sub>2</sub>O, [85]. This was not considered or realized in earlier studies of copper materials, using low temperatures e.g. 800-850°C, where the CLOU effect is small. Low temperatures were used to avoid agglomerations, which were common in laboratory testing at higher temperatures because these normally involved reduction all the way to Cu, which has a low melting temperature, 1079°C.

After the potential advantages with CLOU were realized, most studies have used higher temperatures, and agglomerations have not been noted. This is likely because complete reduction of the oxygen carrier all the way to Cu has been avoided. Both CuO and Cu<sub>2</sub>O have considerably higher melting temperatures as compared to Cu. Operation with methane using higher temperatures has shown excellent gas conversion.

An advantage with copper materials is that the reactions in the fuel reactor are exothermic. Thus, the temperature decrease going from air reactor to fuel reactor, which is inevitable for oxygen carriers with endothermic reactions, can be avoided. Although copper materials have been used in a number of operational studies, there are still uncertainties regarding the material lifetime, [86-88]. Copper materials are costly which means long lifetimes are needed. It should be said that copper oxide impregnated on materials based on  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> has shown low attrition, [89]. Unfortunately, the copper oxide reacts with the support to CuAl<sub>2</sub>O<sub>4</sub>, which is still a good oxygen carrier but without the excellent CLOU properties of CuO, [90-92]. But so far only a limited number of copper oxides have been examined, and the unique performance of copper-CLOU materials makes it worthwhile to pursue the investigation of copper-based materials. Operation with copper-based materials is shown in Table 3.

Table 3: Operation with copper-based oxides.

Operator	Unit	Active oxide/support, prod. method	Time, h	Fuel	Ref.
CSIC	10 kW	CuO/Al <sub>2</sub> O <sub>4</sub> 14/86, WI	120	NG	[93] [94]
Chalmers	0.3 kW	CuO/ZrO <sub>2</sub> 40/60, SD CuO/ZrO <sub>2</sub> -Y 40/60, SD CuO/CeO 40/60, SD	23	NG	[87]
CSIC	0.5 kW	CuO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 14/86, IWI	40	SG	[95]
CSIC	0.5 kW	CuO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 14/86, IWI	30	LHC	[96]
CSIC	0.5 kW	CuO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 14/86, IWI	60	CH <sub>4</sub>	[90]
CSIC	0.5 kW	CuO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 14/86, IWI CuO/ $\alpha$ -Al <sub>2</sub> O <sub>3</sub> 15/85, IWI CuO/MgAl <sub>2</sub> O <sub>4</sub> 12/88, IWI CuO/ $\alpha$ -Al <sub>2</sub> O <sub>3</sub> /NiO 13/84/3, IWI	176	CH <sub>4</sub>	[97]
CSIC	0.5 kW	CuO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 14/86, IWI	40	Sour gas	[98]
CSIC	0.5 kW	CuO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 14/86, IWI	23	Acid gas	[99]
CSIC	0.5 kW	CuO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 14/86, IWI	125	CH <sub>4</sub>	[89]
CSIC	0.5 kW	CuO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 14/86, IWI	65	CH <sub>4</sub>	[100]
CSIC	1 kW-LF	CuO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 14/86, IWI	27	ethanol	[54]
CSIC	0.5/1 kW-LF	CuO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 14/86, IWI	50	Diesel, oil	[58]
Vienna UT	140 kW	CuO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> +CuAl <sub>2</sub> O <sub>4</sub> 9/91, IWI	10	syn	[101]
Vienna UT	140 kW	CuO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 14/86 DI	70	NG	[102] [103]
TU-Vienna	120 kW	CuO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 14/86 DI	50	NG, (H <sub>2</sub> S)	[104]
CSIC	0.5/1.5 kW-SF	CuO/MgAl <sub>2</sub> O <sub>4</sub> 60/40, SD	18	coal	[105]
CSIC	0.5/1.5 kW-SF	CuO/MgAl <sub>2</sub> O <sub>4</sub> 60/40, SD	40	coal	[86]
CSIC	0.5/1.5 kW-SF	CuO/MgAl <sub>2</sub> O <sub>4</sub> 60/40, SD	40	coals	[106]
CSIC	0.5/1.5 kW-SF	CuO/MgAl <sub>2</sub> O <sub>4</sub> 60/40, SD	15	lignite	[107]
CSIC	0.5/1.5 kW-SF	CuO/MgAl <sub>2</sub> O <sub>4</sub> 60/40, SD	10	biomass	[108]
CSIC	0.5/1.5 kW-SF	CuO/Fe <sub>2</sub> O <sub>3</sub> /MgAl <sub>2</sub> O <sub>4</sub> 50/10/40, SD	35	lignite	[109]
CSIC	0.5/1.5 kW-SF	CuO/MgAl <sub>2</sub> O <sub>4</sub> 60/40, SD	10	olive stone, sawdust, almond shell	[110]
Chalmers	0.3 kW LF	CuO/ZrO <sub>2</sub> (Mg) 20/80, SD	45	kerosene	[88]
WKentuU	10 kW	Cu-based	24	NG, syn	[111]

IFP	10 kW	Cu-based	270	CH <sub>4</sub>	[112] [92]
SINTEF	150 kW	CuO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub> 14/86 DI	4	CH <sub>4</sub>	[113]
Hamburg	25 kW –SF	CuO/ $\gamma$ -Al <sub>2</sub> O <sub>3</sub>	5	hard wood, lignite	[114]

#### 2.1.4 Mn-based materials

The oxidized form is Mn<sub>3</sub>O<sub>4</sub> and the reduced form is MnO. MnO cannot be further reduced, not even with very high concentration of reducing gas. Thus, in contrast to the other monometallic oxygen carriers the metallic form will never occur. Thermodynamic calculations show manganese materials could be possible CLOU materials. The oxidized and reduced forms would then be Mn<sub>2</sub>O<sub>3</sub> and Mn<sub>3</sub>O<sub>4</sub>. Unfortunately, the air reactor would need to be at a temperature lower than 800°C to be able to oxidize this material at outlet oxygen concentration of e.g. 5%. In practice, it has not been possible to accomplish the oxidation to Mn<sub>2</sub>O<sub>3</sub> at these temperatures.

Despite the fairly high reactivity and the moderate cost, manganese materials have generally received little attention, and Mn is less studied than Ni, Cu and Fe. Thus, only a few manufactured manganese materials have been used in operation. Operation with manganese materials have shown very high reactivity with CO and H<sub>2</sub>, as well as reasonable reactivity with methane. Manganese materials also appears to be the least likely to form agglomerates, such as has been seen at times with both iron, copper and nickel materials, which is possibly associated with the fact that metallic Mn never forms. Operation with manufactured Mn-based materials is shown in Table 4. Operational data are also available for combined manganese oxides as well as for manganese ores, see subsequent sections.

Table 4: Operation with manufactured manganese-based oxides.

Operator	Unit	Active oxide/support, prod. method	Time, h	Fuel	Ref.
Chalmers	0.3 kW	Mn <sub>3</sub> O <sub>4</sub> /ZrO <sub>2</sub> -Mg 40/60, FG	70	NG/SG	[115]
Chalmers	0.3 kW	Mn <sub>2</sub> O <sub>3</sub> /Fe <sub>2</sub> O <sub>3</sub> 33/67, SD	4	NG	[116]
Chalmers	0.3 kW LF	Mn <sub>3</sub> O <sub>4</sub> /ZrO <sub>2</sub> (Mg) 40/60, SD	17	kerosene	[88]

#### 2.1.5 Fe-based materials

The oxidized form is Fe<sub>2</sub>O<sub>3</sub>, whereas the reduced form is Fe<sub>3</sub>O<sub>4</sub>. Further reduction to FeO or even Fe, is possible, but reduction to these lower states of oxidation is not thermodynamically possible under conditions of full fuel conversion, at least not at well-mixed conditions. However, formation of FeO or Fe locally, where fuel concentration is high, cannot be excluded, although these would be re-oxidized in other zones. In processes for direct hydrogen production, these lower forms are desired, which can be accomplished by designing a fuel reactor where the fuel and oxygen carrier are in counter-current, [117],[118].

Manufactured iron materials generally show poor reactivity towards methane, whereas the reactivity towards syngas is reasonable. Reported operation with manufactured iron oxides is shown in Table 5.

Table 5: Operation with manufactured iron-based oxides.

Operator	Unit	Active oxide/support, prod. method	Time, h	Fuel	Ref.
Chalmers	10 kW	Fe <sub>2</sub> O <sub>3</sub> /MgAl <sub>2</sub> O <sub>3</sub> 60/40, FG	17	NG	[37]
Chalmers	0.3 kW	Fe <sub>2</sub> O <sub>3</sub> /ZrO <sub>2</sub> -Mg 40/60, FG	40	NG/SG	[119]

CSIC	0.5 kW	Fe <sub>2</sub> O <sub>3</sub> /γ-Al <sub>2</sub> O <sub>3</sub> 15/85, WI	40	PSA-OG, CH <sub>4</sub>	[120]
CSIC	0.5 kW	Fe <sub>2</sub> O <sub>3</sub> /NiO/γ-Al <sub>2</sub> O <sub>3</sub> 15/2/83, WI Fe <sub>2</sub> O <sub>3</sub> /γ-Al <sub>2</sub> O <sub>3</sub> 15/85, WI plus NiO/γ-Al <sub>2</sub> O <sub>3</sub> 18/82, WI	32	PSA-OG, CH <sub>4</sub>	[121]
CSIC	0.5 kW	Fe <sub>2</sub> O <sub>3</sub> /γ-Al <sub>2</sub> O <sub>3</sub> 15/85, WI	54	CH <sub>4</sub> /H <sub>2</sub> S	[122]
CSIC	0.5 kW	Fe <sub>2</sub> O <sub>3</sub> /γ-Al <sub>2</sub> O <sub>3</sub> 15/85, WI	20	Sour gas	[98]
CSIC	0.5 kW	Fe <sub>2</sub> O <sub>3</sub> /γ-Al <sub>2</sub> O <sub>3</sub> 15/85, WI	18	Acid gas	[99]
CSIC	1 kW-LF	Fe <sub>2</sub> O <sub>3</sub> /γ-Al <sub>2</sub> O <sub>3</sub> 20/80, WI	27	ethanol	[54]
CSIC	0.5/1 kW-LF	Fe <sub>2</sub> O <sub>3</sub> /γ-Al <sub>2</sub> O <sub>3</sub> 20/80, WI	50	Diesel, oil	[123]
KAIST	1 kW	NiO/Fe <sub>2</sub> O <sub>3</sub> /bent. 15/45/40 Fe <sub>2</sub> O <sub>3</sub> /bentonite 60/40	2	CH <sub>4</sub>	[61]
Vienna UT	140 kW	Fe <sub>2</sub> O <sub>3</sub> /γ-Al <sub>2</sub> O <sub>3</sub> 15/85, WI	34	NG	[124] [103]
Nanjing	10 kW –SF	Fe <sub>2</sub> O <sub>3</sub> SIP	30	biomass	[125]
Ohio	25 kW	supported Fe <sub>2</sub> O <sub>3</sub>	>300 SCL	CH <sub>4</sub>	[126] [127] [128]
Ohio	25 kW –SF	supported Fe <sub>2</sub> O <sub>3</sub>	680 CDLC	coal	[129] [130] [131] [132]
Guangzhou	10 kW-G	Fe <sub>2</sub> O <sub>3</sub> /Al <sub>2</sub> O <sub>3</sub> 70/30	60	saw dust	[133] [134]
Guangzhou	10 kW-G	Fe <sub>2</sub> O <sub>3</sub> /Al <sub>2</sub> O <sub>3</sub> /NiO 7/3/0.53	2	saw dust	[135]
NCCC	250 kW	Fe <sub>2</sub> O <sub>3</sub> -based	360	SG+propane	[118]

### 2.1.6 Cobalt oxide CoO

The oxidized and reduced forms of this system are CoO and Co. For good reason little work has been done with cobalt-based materials. It is even more toxic and more costly than Ni materials and thermodynamic restraints prevent conversion of methane to CO<sub>2</sub> to 95-97%. However, thermodynamics indicate that cobalt could be used for CLOU, using the system Co<sub>3</sub>O<sub>4</sub>/CoO, but to oxidize CoO to Co<sub>3</sub>O<sub>4</sub> would require temperatures below 845°C in 4% oxygen, [26].

Table 6. Operation with cobalt-based oxides.

Operator	Unit	Active oxide/support, prod. method	Time, h	Fuel	Ref.
KIER	50 kW	Co <sub>x</sub> O <sub>y</sub> /CoAl <sub>2</sub> O <sub>4</sub> 70/30, CP/I	28	NG	[136]
KIER	50 new kW	<sup>1</sup> / <sub>3</sub> NiO/bentonite 60/40, MDCC + <sup>2</sup> / <sub>3</sub> Co <sub>x</sub> O <sub>y</sub> /CoAl <sub>2</sub> O <sub>4</sub> 70/30, CP/I	100-200	nat.gas, SG	[78]

### 2.1.7 Combined oxide materials

Combined metal oxides, i.e. where two or more oxides are combined not only physically, but chemically, constituting new oxides, for example calcium manganite, CaMnO<sub>3-δ</sub>. Combined Mn oxides may exhibit CLOU properties, i.e. the ability to release oxygen. Such materials include Mn combined with Ca, Fe, Si, Mg, Cu and Ni. The thermodynamic properties of these combined manganese oxides were investigated by Rydén et al., [27].

Operation with combined oxides is shown in Table 7. Much of the operation involves various calcium manganites, which have a perovskite structure. Although these materials have a lower direct reactivity towards methane than nickel materials, they seem to be able to perform equally, or even better in pilot operation. The reason is likely that the release of oxygen makes it possible to convert methane which is not in direct contact with the oxygen carrier. Thus, the

by-pass of gas in fluidized beds should be less cumbersome with a CLOU material. If temperature and circulation are sufficient and fuel load not too high, operation with calcium manganite has shown full conversion and even an excess of oxygen in operation with natural gas, [137]. With high volatile solid fuel, i.e. biomass, a dramatic improvement in gas conversion compared to natural ores was observed,[138].

Table 7. Operation with manufactured combined oxides.

Operator	Unit	Active oxide/support, prod. method	Time, h	Fuel	Ref.
Chalmers	10 kW	CaMn <sub>0.9</sub> Mg <sub>0.1</sub> O <sub>3</sub> , SD	55	NG	[137]
Chalmers	10 kW	CaMn <sub>0.78</sub> Mg <sub>0.1</sub> Ti <sub>0.12</sub> O <sub>3</sub> , SD	99	NG	[139]
Chalmers	10 kW	CaMn <sub>0.78</sub> Mg <sub>0.1</sub> Ti <sub>0.12</sub> O <sub>3</sub> , SD (E1S2, 901)	134	NG	[140]
Chalmers	0.3 kW	CaMn <sub>0.87</sub> Ti <sub>0.13</sub> O <sub>3</sub> , SP+FG	70	NG	[141]
Chalmers	0.3 kW	(Mn <sub>0.3</sub> Fe <sub>0.5</sub> )TiO <sub>3</sub>	12	NG	[142]
Chalmers	0.3 kW	FeMnSiO <sub>3</sub> , SD Fe <sub>0.66</sub> Mn <sub>1.33</sub> SiO <sub>3</sub> , SD	8+16	NG	[143]
Chalmers	0.3 kW	CaMn <sub>0.8</sub> Mg <sub>0.2</sub> O <sub>3</sub> , SD CaMn <sub>0.9</sub> Mg <sub>0.1</sub> O <sub>3</sub> , SD CaMn <sub>0.78</sub> Mg <sub>0.1</sub> Ti <sub>0.12</sub> O <sub>3</sub> , SD	15+16+ 40 =71	NG	[144]
Chalmers	0.3 kW	Calcium manganite SD	35	NG	[145]
Chalmers	0.3 kW	<sup>2</sup> / <sub>3</sub> (Fe <sub>0.33</sub> Mn <sub>0.67</sub> )O <sub>3</sub> <sup>1</sup> / <sub>3</sub> SiO <sub>2</sub> <sup>7</sup> / <sub>9</sub> (Fe <sub>0.29</sub> Mn <sub>0.71</sub> )O <sub>3</sub> <sup>2</sup> / <sub>9</sub> SiO <sub>2</sub> <sup>8</sup> / <sub>9</sub> (Fe <sub>0.25</sub> Mn <sub>0.75</sub> )O <sub>3</sub> <sup>1</sup> / <sub>9</sub> SiO <sub>2</sub>	10+11+ 14 =35	NG	[146]
Chalmers	0.3 kW	Mn <sub>3</sub> O <sub>4</sub> /SiO <sub>2</sub> (75/25) Mn <sub>3</sub> O <sub>4</sub> /SiO <sub>2</sub> /TiO <sub>2</sub> (67/22/11) SD	7 + 24 =31	NG, syn	[147]
Chalmers	0.3 kW	Mn <sub>3</sub> O <sub>4</sub> /Fe <sub>2</sub> O <sub>3</sub> /Al <sub>2</sub> O <sub>3</sub> Mn:Fe:Al = 1:2:0.64	26	syn, CH <sub>4</sub> , kerosene	[148]
Chalmers	10 Kw SF	CaMn <sub>0.9</sub> Mg <sub>0.1</sub> O <sub>3</sub> , SD	74	wood char, petcoke	[149] [150]
Chalmers	10 kW SF	Mn <sub>3</sub> O <sub>4</sub> /SiO <sub>2</sub> /TiO <sub>2</sub> (67/22/11) SD	32	w-char, coal, petcoke, lign	[151]
CSIC	0.5 kW	CaMn <sub>0.9</sub> Mg <sub>0.1</sub> O <sub>3</sub> , SD	71	CH <sub>4</sub> /H <sub>2</sub> S	[152]
CSIC	0.5 kW	(Mn <sub>0.77</sub> Fe <sub>0.23</sub> ) <sub>2</sub> O <sub>3</sub>	10	CH <sub>4</sub> , syn	[153]
Vienna UT	140 kW	CaMn <sub>0.9</sub> Mg <sub>0.1</sub> O <sub>3</sub> , SD	30	NG	[124] [103]
Vienna UT	140 kW	CaMn <sub>0.78</sub> Mg <sub>0.1</sub> Ti <sub>0.12</sub> O <sub>3</sub> , SD	23	NG	[124] [103]
Vienna UT	140 kW	CaMn <sub>0.78</sub> Mg <sub>0.1</sub> Ti <sub>0.12</sub> O <sub>3</sub> , SD (ES)	11	NG (+ H <sub>2</sub> S)	[154]
Xi'an Jiaotong	10 kW- Pr	Fe <sub>2</sub> O <sub>3</sub> /CuO/MgAl <sub>2</sub> O <sub>4</sub> 45/15/40, Ext	15	coke oven gas\	[155]
Darmstadt	1 MW	CaMn <sub>0.78</sub> Mg <sub>0.1</sub> Ti <sub>0.12</sub> O <sub>3</sub> , SD	60	NG	[156] [157]
Darmstadt	1 MW	CaMn <sub>0.78</sub> Mg <sub>0.1</sub> Ti <sub>0.12</sub> O <sub>3</sub> , SD + ilmenite	20	NG	[157]
NETL	50 kW	CuO/Fe <sub>2</sub> O <sub>3</sub> /Al <sub>2</sub> O <sub>3</sub> 40/30/30	40	NG	[158, 159]
NETL	50 kW	CuO/Fe <sub>2</sub> O <sub>3</sub> /Al <sub>2</sub> O <sub>3</sub> WG	40	CH <sub>4</sub>	[160]
NETL	50 kW	CuO/Fe <sub>2</sub> O <sub>3</sub> /Al <sub>2</sub> O <sub>3</sub> WG	11	CH <sub>4</sub>	[161]
Chalmers	10 kW LF	CaMn <sub>0.9</sub> Mg <sub>0.1</sub> O <sub>3</sub> , SD	10	Heavy fuel oil	[162]
CSIC	0.5/1.5 kW-SF	CuO/Fe <sub>2</sub> O <sub>3</sub> /MgAl <sub>2</sub> O <sub>4</sub> 50/10/40, SD	35	lignite	[109]
CSIC	0.5/1.5 kW-SF	Cu <sub>34</sub> Mn <sub>66</sub> , SG	40	olive stone, sawdust, almond shell	[110]
CSIC	0.5/1.5 kW-SF	(Mn <sub>0.77</sub> Fe <sub>0.23</sub> ) <sub>2</sub> O <sub>3</sub>	10	4 coals	[153]
IFP	10 kW	CaMn <sub>0.78</sub> Mg <sub>0.1</sub> Ti <sub>0.12</sub> O <sub>3</sub> , SD (ES)	75	CH <sub>4</sub>	[163]
Chalmers	100 kW- SF	CaMn <sub>0.78</sub> Mg <sub>0.1</sub> Ti <sub>0.12</sub> O <sub>3</sub> , SD (ES) + ilmenite	18	biomass	[164] [138]

### 2.1.8 Fe-based low-cost materials

Early studies of iron ores showed low reactivity towards methane, [165], but the low price of iron ores in combination with reasonable reactivity towards syngas, make iron ores an interesting option for chemical-looping combustion of solid fuels. Table 8 shows an overview of operation with iron ores and iron oxide waste materials.

Table 8. Operation with natural/waste iron oxides materials.

Operator	Unit	Oxygen carrier*	Time, h	Fuel	Ref.
Chalmers	0.3 kW	Fe <sub>2</sub> O <sub>3</sub> shells	37	SG	[166]
Chalmers	0.3 kW	Steel slag (Ca, Fe, Si, Mg, Mn)	20	SG, CH <sub>4</sub>	[167]
CSIC	0.5 kW	Fe <sub>2</sub> O <sub>3</sub> WM	111	CH <sub>4</sub> SG	[168]
CSIC	0.5 kW	iron ore (Es)*	50	CH <sub>4</sub> , SG, PSA off-gas	[169]
Nanjing	1 kW – SF	iron ore (Au)	10	coal	[170]
Nanjing	1 kW – SF	iron ore (Au)	?	sawdust/coal	[171]
Nanjing	1 kW – SF	iron ore (Au) + 3% of NiO/NiAl <sub>2</sub> O <sub>4</sub> /Al <sub>2</sub> O <sub>3</sub> 20/39/41, CP	10	coal	[172]
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	[173]
Nanjing	1 kW – SF	iron ore	20	coal/anthracite	[174]
Nanjing	1 kW – SF	iron ore + K	5	coal	[175]
Nanjing	1 kW – SF	iron ore	22	coal with high K	[176]
Nanjing	1 kW – SF	iron ore	10	sewage sludge	[177]
Nanjing	1 kW – SF	iron ore + cement/CaO	15	coal	[178] [179]
Nanjing	1 kW – SF	iron ore	5	coal	[180]
CSIC	0.5/1.5 kW-SF	Fe WM	40	coal	[181]
CSIC	0.5/1.5 kW-SF	Fe ore (Es)	78	biomass	[182]
CSIC	0.5/1.5 kW-SF	Fe ore (Es)	30	coals	[183]
CSIC	0.5/1.5 kW-SF	Fe ore (Es)	18	anthracite, lignite	[184]
CSIC	0.5/1.5 kW-SF	Fe ore (Es)	40	saw dust, olive stone, almond shell	[185] [186]
Chalmers	100 kW-SF	Fe ore (Es)	26	wood char, 2 coals	[187]
Huazhong	5 kW –G/SF	Fe ore	100	CH <sub>4</sub>	[188]
Huazhong	5 kW – G/SF	Fe ore	100	coal	[189]
Huazhong	50 kW –SF	Fe ore	6	coal	[190]
Nanjing	25 kW-SF	Fe Ore	>6	rice husk	[191]
Nanjing	50 kW-Pr SF	Fe Ore	19	coal	[192]
Nanjing	20 kW-SF	Fe Ore	70	coal	[193]
Huazhong	50 kW –SF	Fe ore	2	coal	[194]
Huazhong	50 kW –SF	Fe ore	6	coal	[190]
Zabrze	10 kW	Fe ore	3	CH <sub>4</sub>	[195]
NETL	50 kW	iron ore	2	NG	[196]
CSIC	50 kW-SF	iron ore	20	olive stone, saw dust	[197]
CSIC	50 kW-SF	iron ore	15	coal	[198]
Chalmers	10 kW–SF	Steel slag (Ca, Fe, Si, Mg, Mn)	28	wood char, wood pellets	[167]
Darmstadt	1 MW	iron ore	42	coal, torrif. biomass	[199]
Nanjing	5 kW-SF/s	iron ore	8	sewage sludge	[200]
Nanjing	5 kW-SF/s	iron ore	4	gasification of coal	[201]
Nanjing	5 kW-SF/s	iron ore	4	coal	[202]
Nanjing	5 kW-SF/i	iron ore	3	biomass	[203]

Nanjing	5 kW-SF/i	iron ore	3	CO	[203]
Nanjing	2 kW	iron ore	12	SG, NG	[204]
Nanjing	25 kW	iron ore	2	coal	[205]

\*Abbreviations in parenthesis after ores indicate country of origin.

### 2.1.9 Ilmenite

The combined oxide ilmenite,  $\text{FeTiO}_3$ , is a naturally occurring mineral.  $\text{FeTiO}_3$  is the reduced form and the oxidized form is  $\text{Fe}_2\text{TiO}_5 + \text{TiO}_2$ , [206]. In operation there is a phase separation with migration of Fe to the surface, giving an outer  $\text{Fe}_2\text{O}_3$  layer, which will also participate in the oxygen transfer. The important advantage of ilmenite is the low price in combination with a reasonable reactivity towards syngas. Table 9 shows operation with ilmenite materials.

Table 9. Operation with ilmenite ore.

Operator	Unit	Oxygen carrier	Time, h	Fuel	Ref.
Chalmers	0.3 kW	ilmenite (No) w Ni	83	NG	[207]
Chalmers	0.3 kW	ilmenite (No)	85	SG	[166]
Chalmers	10 kW-SF	ilmenite (No)	22	coal	[208]
Chalmers	10 kW-SF	ilmenite (No)	11	petcoke	[209]
Chalmers	10 kW-SF	ilmenite (No)	18	petcoke	[210]
Chalmers	10 kW-SF	ilmenite (No)	26	petcoke	[211]
Chalmers	10 kW-SF	ilmenite (No)	4	petcoke	[212]
Chalmers	10 kW-SF	ilmenite (No) + lime	4	petcoke	[212]
Chalmers	10 kW-SF	ilmenite (No)	29	petcoke, coal	[213]
Vienna UT	140 kW	ilmenite (No)	appr. 160*	$\text{CH}_4$ , CO, $\text{H}_2$	[214] [68] [69]
Stuttgart	10 kW	ilmenite (Au)	1	SG	[215]
CSIC	0.5/1.5 kW-SF	ilmenite (No)	26	coal	[216]
CSIC	0.5/1.5 kW-SF	ilmenite (No)	35	coal	[217]
CSIC	0.5/1.5 kW-SF	ilmenite (No)	30	coal	[218]
CSIC	0.5/1.5 kW-SF	ilmenite (No)	44	coals	[219]
CSIC	0.5/1.5 kW-SF	ilmenite (No)	35	lignite	[220]
Chalmers	0.3 kW LF	ilmenite (No)	80	kerosene	[221]
Chalmers	100 kW-SF	ilmenite (No)	24	coal, PC	[222] [223] [224] [225]
Chalmers	100 kW-SF	ilmenite (No)	12	wood char	[226]
Chalmers	100 kW-SF	ilmenite (No)	34	wood char, PC	[227]
Chalmers	100 kW-SF	ilmenite (No) + Mn ore	18	wood char, 2 PC	[228]
Hamburg	25 kW -SF	ilmenite (Au)	>60	$\text{CH}_4$ , coal (21 h)	[229]
Hamburg	25 kW -SF	ilmenite (Au)	30	coal, lignite, biomass	[230]
Chalmers	10 kW LF	Ilmenite (No)	66	Diesel	[231]
Chalmers	10 kW LF	Ilmenite (No)	4	Heavy fuel oil	[162]
CSIC	50 kW-SF	Ilmenite (No)	4	coal	[232]
CSIC	50 kW-SF	Ilmenite (No)	30	coal	[233]
Darmstadt	1 MW	Ilmenite (No)	2	coal	[234]
Darmstadt	1 MW	Ilmenite (No)	3	coal	[235]
Darmstadt	1 MW	Ilmenite (No)	12	coal	[199]
Darmstadt	1 MW	Ilmenite + iron ore	56	coal	[199]
Tsinghua	0.2 kW	Ilmenite	140	CO	[236]
Chalmers	1.4/10 MW	Ilmenite	61	biomass	[237]
VTT	20 kW	Ilmenite (No)	16	biomass	[238]
SINTEF	150 kW	Ilmenite (No)	5	wood pellets	[239]
CSIRO	10 kW-SF	Ilmenite (Au)	35	brown coal	[240]
Tsinghua	30 kW-SF	Ilmenite (Vn)	100	coal	[241]
JCOAL	100 kW-GSF	Ilmenite	64	NG	[242]
JCOAL	100 kW-GSF	Ilmenite	7	coal	[242]
Vienna	80 kW-SF	Ilmenite (No)	20	wood pellets	[243]

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### 2.1.10 Mn-based low-cost materials

Manganese ores are not as cheap as iron ores, but they still have low cost. The oxidation state of manganese in the ores vary and often the manganese combines with other elements to form various minerals. Because Si and Fe are normally present in manganese ores, these could also potentially have CLOU properties. Laboratory testing indicates that most manganese ores have the ability to release at least minor amounts of oxygen, [244] [245]. Operation with manganese ores generally shows better conversion but more dust formation as compared to ilmenite. Operation with manganese ores is shown in Table 10.

Table 10. Operation with manganese ores.

Operator	Unit	Oxygen carrier	Time, h	Fuel	Ref.
Chalmers	0.3 kW	Mn ore	2	NG	[116]
Chalmers	0.3 kW	5 Mn ores	111	NG, syn, vol	[246]
Chalmers	0.3 kW	Mn ore	21	syn, CH <sub>4</sub> , kerosene	[148]
Chalmers	10 kW-SF	Mn ore (Br)	10	petcoke	[213]
Chalmers	10 kW-SF	Mn ore (Br) + lime	15	petcoke	[247]
Chalmers	10 kW-SF	Mn ore Mn ore Mn ore	16+15 +11 =42	wood char, pet coke	[248]
Chalmers	10 kW-SF	Mn ore	22	wood char, wood pellets, coal	[249]
IFP-Lyon	10 kW-GSF	Mn ore	38	CH <sub>4</sub> , SG	[250]
IFP-Lyon	10 kW-GSF	Mn ore	52	coal	[250]
Tsinghua	1 kW	Mn ore, Mn ore + Cu	182	CO	[251]
Chalmers	100 kW-SF	Mn ore	52	w-char, coals, petcoke, lign.	[252]
Chalmers	100 kW-SF	Mn ore	33	wood char, wood pellets, coal	[249]
VTT	20 kW	Mn ore	23	biomass	[253]
Chalmers	1.4/10 MW	Mn ore	32	biomass	[254]
CSIC	0.5 kW-SF	Mn ore (Ga)	100	coal	[255]

### 2.1.11 Other low-cost materials

Limestone is a cheap and abundant material that can be sulphated to form CaSO<sub>4</sub>. CaSO<sub>4</sub>/CaS has been studied as a low cost oxygen carrier for solid fuels. It has a uniquely high oxygen transfer capacity, 47%, but it has a thermodynamic constraint and cannot convert CO and H<sub>2</sub> more than 98-99%. There is also a risk of sulphur being lost, converting the oxygen carrier to CaO. Loss of sulphur is difficult to predict as it takes place in the shifts between oxidizing and reducing conditions and will be very dependent on the process conditions, including temperature, fuel sulphur content, extent of fuel conversion and frequency of shifts, [256-258]. Operation with the CaSO<sub>4</sub>/CaS system is shown in Table 11.

Table 11. Operation with other low-cost materials.

Operator	Unit	Active oxide/support, prod. method	Time, h	Fuel	Ref.
Alstom	3 MW-SF	CaSO <sub>4</sub> (FBC ash)	75	coal	[259]

## 2.2 Chemical-looping combustors

Since the first operation was reported in 2004, data from a rising number of chemical-looping combustors have been published, see Fig. 5. Further, the fraction of solid fuel combustors has increased from zero to 60%. Moreover, as indicated by the trend in Fig. 6, the chemical-looping combustors have generally become larger.

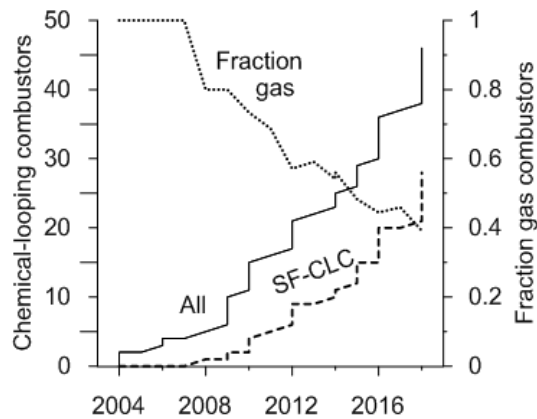


Figure 5. Number of chemical-looping combustors versus year

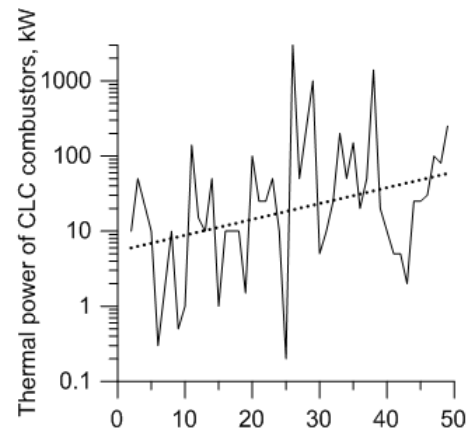


Figure 6. Size of chemical-looping combustors versus consecutive number.

Table 12 lists the 46 chemical-looping combustors used in operation and Table 13 explains the short names for the 25 operators. All these pilots, except those at Ohio State University and NCCC, use interconnected fluidized beds. In Ohio and at NCCC the fuel reactor is a moving bed.

A majority of the operation reported is from Europe, 70%, see Fig. 7, and is associated with a number of European projects that involved Chalmers, CSIC, Vienna, as well as Alstom, Darmstadt, IFP and SINTEF. In Asia an important part of the operation is with Nanjing and KIER, and in North America Ohio State University dominates.

Table 12. Operation of 46 chemical-looping combustors/ gasifiers.

Operator	Unit	Hours of operation	Typical fuels used, selected references	First reported
1 Chalmers	10 kW	1570	nat. gas [36] [37]	2004
2 KIER	50 kW	31	nat. gas [40]	2004
3 CSIC	10 kW	120	nat. gas [93]	2006
4 Chalmers	0.3 kW-GL	1359	nat. gas, syngas, kerosene [42]	2006
5 Chalmers	10 kW-SF	337	coal, petcoke, biomass pellets, wood char [260] [208]	2008
6 CSIC	0.5 kW-GL	1812	nat. gas, acid gas, sour gas, ethanol [47]	2009

7	KAIST	1 kW	8 CH <sub>4</sub> [61]	2009
8	Vienna UT	140 kW	660 nat. gas, CO, H <sub>2</sub> [67]	2009
9	Alstom, Fr	15 kW	100 nat. gas [74]	2009
10	Nanjing	10 kW –SF	260 coal, biomass. [261]	2009
11	KIER	50 kW	300 nat.gas, syngas [78]	2010
12	Nanjing	1 kW – SF	195 coal, biomass, sew. sludge [170] [171]	2010
13	IFP-Lyon	10 kW-GSF	453 CH <sub>4</sub> , coal, syngas [262] [250]	2010
14	Stuttgart	10 kW	1 syngas [215]	2010
15	Xi'an Jiaotong	10 kW- Pr	15 coke oven gas [155]	2010
16	CSIC	1.5 kW-SF	729 coal [217]	2011
17	Chalmers	100 kW – SF	217 coal, petcoke, wood pellets, wood char [223] [224]	2012
18	Hamburg	25 kW –SF	95 coal, CH <sub>4</sub> [263]	2012
19	Ohio	25 kW –SF	980 coal [127] [264]	2012
20	Nanjing	50 kW-Pr	19 coal [192]	2012
21	WKentuU	10 kW	24 nat. gas, syngas [111]	2012
22	Tsinghua	0.2 kW	322 CO [236]	2013
23	Alstom, US	3 MW –SF	75 coal [259]	2014
24	CSIC	50 kW-SF	69 coal, lignite, anthracite [232]	2014
25	Chalmers	10 kW-LF	80 diesel, heavy fuel oil [162]	2014
26	Darmstadt	1 MW –GSF	195 coal [234] [235]	2015
27	Huazhong	5 kW-GSF	200 CH <sub>4</sub> coal [189]	2015
28	Guangzhou	10 kW-G	62 saw dust [133]	2015
29	Nanjing	25 kW-G	13 rice husk [83]	2015
30	KIER	200 kW	100 nat. gas [84]	2016
31	Huazhong	50 kW-SF	8 coal [194]	2016
32	SINTEF	150 kW	9 CH <sub>4</sub> , biomass [113]	2016
33	VTT	20 kW-SF	130 biomass [238]	2016
34	NETL	50 kW	2 CH <sub>4</sub> [265]	2016
35	Chalmers	1.4/10 MW	93 biomass [237]	2016
36	Nanjing	20 kW-SF	70 coal [193]	2016
37	Zabrze	10 kW	3 CH <sub>4</sub> [195]	2017
38	Nanjing	5 kW-SF/s	16 coal, sewage sludge [200]	2017
39	Nanjing	5 kW-SF/i	6 biomass, CO, [203]	2018
40	Nanjing	2 kW-SF	12 syngas, nat. gas [204]	2018
41	Nanjing	25 kW-G	2 coal [205]	2018
42	CSIRO	25 kW-SF	35 brown coal [240]	2018
43	Tsinghua	30 kW-SF	100 coal [241]	2018
44	JCOAL	100 kW-GSF	73 NG, coal [242]	2018
45	Vienna UT	80 kW-SF	20 wood pellets [243]	2018
46	NCCC	250 kW Pr WS	360 syngas+propane [118]	2018

SF-solid fuel, GSF-gaseous & solid fuel, Pr-pressurized, LF-liquid fuel, GL=gaseous/liquid fuel, G-Gasification, WS=water splitting, /s=staged, /i=with internals

Table 13. Operators of CLC pilots.

Alstom, Fr	Alstom Power Boilers, France
Alstom, US	Alstom Power Inc., Windsor, US (now GE)
Chalmers	Chalmers University of Technology, Gothenburg, Sweden
CSIC	Consejo Superior de Investigaciones Científicas, Instituto de Carboquímica, Zaragoza, Spain
CSIRO	Commonwealth Scientific and Industrial Research Organisation, Clayton South, Australia
Darmstadt	Darmstadt University of Technology, Germany

Guangzhou	Guangzhou Institute of Energy Conversion, China
Hamburg	Technical University Hamburg-Harburg, Germany
Huazhong	Huazhong University of Science and Technology, China
IFP-Lyon	Institut Francais du Petrole, Lyon, France
KAIST	Korea Advanced Institute of Science and Technology, Daejeon, Korea
JCOAL	Japan Coal Energy Center, Tokyo, Japan
KIER	Korea Institute of Energy Research, Daejeon, Korea
Nanjing	South East University, Nanjing, China
NCCC	National Carbon Capture Centre, Wilsonville, Alabama
NETL	National Energy Technology Laboratory, US
Ohio	Ohio State University, US
SINTEF	SINTEF Energy Research, Trondheim, Norway
Stuttgart	University of Stuttgart, Germany
Tsinghua	Tsinghua University, Beijing, China
Vienna UT	Vienna University of Technology, Austria
VTT	VTT Technical Research Centre of Finland
WKentuU	Western Kentucky University, Bowling Green, US
Xi'an Jiaotong	Xi'an Jiaotong University, Xi'an, China
Zabrze	Institute for Chemical Processing of Coal, Zabrze, Poland

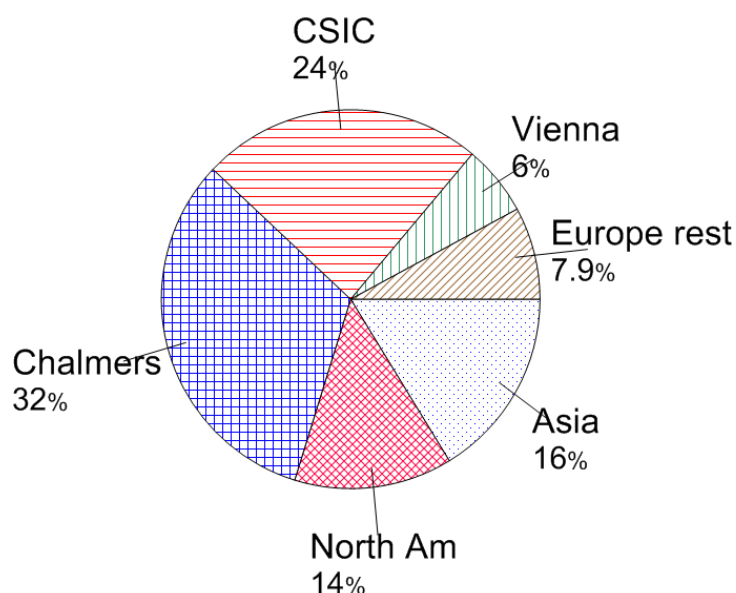


Figure 7. Fraction of total operation versus location.

### 3 Where are we and where are we going?

#### 3.1 Where are we?

When looking at the rapid accumulation of experience from operation of Chemical-Looping Combustion it should be acknowledged that there is a significant gap to close in comparison to established and well-known technologies. In fact, Chemical-Looping Combustion represents a novel principle of fuel oxidation. In contrast to normal combustion using direct contact between oxidizing gas (air) and fuel, chemical-looping is an indirect method using a medium for oxygen transfer, in similarity to respiration, used by the biosphere since 2 billion years, and fuel cells demonstrated first in 1839, Table 14.

Table 14. Principles of hydrocarbon fuel oxidation.

Principle	first demonstration by human beings or living organisms
respiration	~2 000 000 000 B.C
combustion	~500 000 B.C
fuel cells	1839
chemical-looping combustion	2003

Figures 4-5 and Table 1 clearly demonstrate an increasing activity in the chemical-looping research. It should be also remembered that the earlier phase of development was, for good reason, more focused on the low-hanging fruits. As the operational experiences accumulate, the demands for novel information, that may be more difficult to come by, increases. From Fig. 5 and 6, showing the increased use of solid fuels and the increase in unit size, it is clear that the more recent operational experiences have a greater focus on more difficult tasks.

Firstly the operation of solid and liquid fuels is more costly and requires more manpower. Chemical-looping combustion of solid fuels require much more preparatory work and supplementary work, e.g. in order to establish the fate of fuel particles and constituents and its interaction with the oxygen carrier. Even though SF-CLC comes with expectations of dramatic reductions in CO<sub>2</sub> capture costs and energy penalties, it is undeniable that, at least from the point of view of research, it comes with greater complexity and costs.

Secondly, the operation of larger units also comes with similarly increased costs and increased requirements for manpower. Thirdly, there is also a much greater focus on more complex materials, both the combined manufactured materials and the natural ores and waste materials. The natural materials are normally quite complex containing a number of oxides that may make up a number of various combined oxides. As an example the average composition and standard deviation from a comparison of eight natural manganese ores are given in Table 15, showing firstly high concentrations of several oxides that are likely to interact with manganese as well as high standard deviations suggesting that the materials could have different properties.

*Table 15. Average analysis of 8 manganese ores. Data from [245]*

	Average, %	Standard deviation
SiO <sub>2</sub>	11.0	3.5
Al <sub>2</sub> O <sub>3</sub>	4.6	4.2
CaO	5.3	6.5
Fe <sub>2</sub> O <sub>3</sub>	27.7	21.6
K <sub>2</sub> O	0.6	0.4
MgO	2.1	2.6
Mn <sub>3</sub> O <sub>4</sub>	49.5	21.4

Clearly, the research activities are comprehensive, and the scope is expanding leading to rapid accumulation of important knowledge and experience. Important is the increasing operational experiences with an increased number of oxygen-carrier materials. This means there is an increasing portfolio of materials that can be suitable for different applications of chemical-looping technologies, or under different conditions. Economic optimizations, commercial experiences of the technology, technology developments, or other changes in conditions may shift the emphasis on what is actually the best particle properties in relation to expected lifetime, reactivity, price, toxicity and suitable temperature range. A portfolio of different materials is also an important risk reducer for large-scale investments in the technology.

Na <sub>2</sub> O	0.22	0.18
P <sub>2</sub> O <sub>5</sub>	0.19	0.07
TiO <sub>2</sub>	0.29	0.38

### 3.1.1 Oxygen Carrier Aided Combustion (OCAC)

A spin-off from the quest for oxygen carriers for chemical-looping combustion, is the idea to use oxygen carrier materials to improve fluidized-bed combustion, called Oxygen Carrier Aided Combustion (OCAC). OCAC has been investigated in a 12 MW<sub>th</sub> CFB boiler, using ilmenite, [266], and manganese ore, [267], as oxygen carrier. Moreover, ilmenite has been used as bed material in a 75 MW<sub>th</sub> CFB for more than 12,000 h of operation, [268]. The advantages using oxygen-carrier materials are significant and may outweigh an added cost of such bed materials. For chemical-looping combustion the use of oxygen carriers in OCAC has firstly demonstrated that oxygen carriers can be used in large-scale fluidized beds with fuels such as biomass and municipal waste, and has secondly provided the experience with acquisition and handling of oxygen carrier in the scale of a thousand tonnes.

## 3.2 Where are we going?

A number of developments of chemical-looping combustion are going on in parallel. At the same time the rules of the game in which CLC will play are changing and can be expected to change fundamentally. Meeting the Paris climate agreement will require very fundamental changes to the global energy system, which will not be possible without introduction of effective means of governance. We can only guess what this will mean in practice, but a combination of rapidly decreasing fossil CO<sub>2</sub> emissions and atmospheric carbon removal, i.e. negative CO<sub>2</sub> emissions, will be needed. Consequently, several schemes with incentives, regulations and subsidies could come out of this.

A few points with likely bearing on the future development of SF-CLC will be discussed next.

### 3.2.1 What will happen to performances as the process is up-scaled?

A comparison of the performance of the four largest solid-fuel pilots, 50 kW to 3 MW, is given in [20]. The largest SF-CLC units so far have a size of 1 and 3 MW. The latter used CaSO<sub>4</sub>/CaS and no comparison to smaller pilots is available for this system. In case of the 1 MW pilot a significantly lower performance has been noted. However, it is most likely that this can be explained by factors like low fuel reactor solids inventory, high level of fuel inlet, and low fuel reactor temperature, [20]. Conditions in fluidized-beds change fundamentally when scale is increased. Higher velocities can be expected give a larger fraction of the gas by-

passing the bed in bubbles and through-flow, i.e. poorer mass transfer, as long as we remain in the so-called bubbling-bed regime. However, further increase of velocity going into the turbulent and fast fluidization regime can be expected to come with increased mass transfer,[269]. But the hard truth is that it is difficult to safely predict the performance in larger units.

### 3.2.2 Key technology challenges

In general, CLC technology can adapt circulating fluidized bed (CFB) technology, but there are also differences, [20], involving some important challenges, e.g. control of circulation, optimizing gas conversion by design of fuel reactor and selection of oxygen carrier, control of char loss to stack and air reactor by fuel particle size and fuel reactor design, and finally adequate downstream treatment of fuel reactor effluent.

### 3.2.3 Routes for scaling up at reduced costs

An important barrier against the scaling up of the technology, is obviously the cost and, perhaps even more important, the reluctance to risk money on technology not previously demonstrated in full scale. Lyngfelt et al., [20], discusses strategies to significantly reduce the financial risks involved, e.g. using an existing CFB, or construction of a dual purpose CLC/CFB. The dual purpose approach can also be combined with starting a demonstration unit without the costly process steps needed to attain storable CO<sub>2</sub>, i.e. without oxygen production for oxygen polishing and downstream treatment involving purification and compression of the CO<sub>2</sub>. These can be added after successful demonstration of the core process and be optimized for the composition of the effluent stream obtained. Alternatively, the unit can be used as a CFB. A further development of this idea is the concept of a multipurpose dual fluidized-bed (MDFB), with several different potential applications, [270].

### 3.2.4 Costs of solid-fuel CLC

Proposed designs of solid-fuel chemical-looping boilers presented in the literature involve a 100 MW<sub>th</sub> and a 1000 MW<sub>th</sub> unit, [271], [272]. The latter also involves a cost estimation indicating an added cost of CLC as compared to a circulating fluidized bed (CFB) boiler of 16-26 €/tonne of CO<sub>2</sub> captured. The major costs are downstream of the process, i.e. compression and oxygen production for oxy-polishing. Therefore, the two step approach suggested in the previous section could make sense. Other costs presented for SF-CLC of coal are 10 €/tonne, [273], 26 €/tonne, [274], and 32 €/tonne, [275]. The latter found a lower cost for biomass, 24 €/tonne.

Another aspect of the costs is that chemical-looping may come with other advantages, in the case of coal potentially very low emissions of SO<sub>2</sub> and NO<sub>x</sub>. A techno-economic analysis indicates the cost of both SO<sub>2</sub> and NO<sub>x</sub> removal should go down by a factor of three,[276]. Thus, CLC could be a solution when strengthened emission policies go beyond what is reached with CFB technology. As will be discussed in section 3.2.6 chemical-looping combustion could also come with advantages with respect to difficulties associated with alkali compounds in the fuel ash.

In conclusion, SF-CLC shows a unique potential for attaining very significant cost reduction for CO<sub>2</sub> capture. This is not an unexpected result, as the technology avoids very costly gas separation processes.

### *3.2.5 Funding of CLC*

At present, incentives for reducing CO<sub>2</sub> emissions or for negative CO<sub>2</sub> emissions, are generally poor or non-existent. But, as discussed above, this can be expected to change, and there are many possible means of introducing incentives, [277]. As an interesting example, the California Low Carbon Fuel Standard (LCFS), has created a cap-and-trade market for CO<sub>2</sub> credits which will start to include negative emissions from the fall of 2018, [278]. These credits are typically sold at 120 \$/tonne of CO<sub>2</sub>, [279]. To be added to this price is also the possibility of a tax deduction of 50 \$/tonne for negative emissions. In total this could mean 170 \$/tonne of CO<sub>2</sub> for negative emissions. This is ten times more than the lowest estimations of costs for CLC.

## **4 Negative CO<sub>2</sub> emissions using biomass**

### **4.1 Negative CO<sub>2</sub> emissions**

The concept of CO<sub>2</sub> budgets is used to show the total amount of emissions that would still be allowed for meeting climate stabilization targets. The estimated remaining budget for a maximum temperature rise of 2°C is around 700-800 Gt CO<sub>2</sub>, [280]. With present emissions of 35-40 Gt/year, this would be exhausted in approximately 20 years from now. It is evident that such a rapid reduction in CO<sub>2</sub> emissions is not in correspondence with national plans for CO<sub>2</sub> reductions. This is because the modelling scenarios include a back-door, namely negative CO<sub>2</sub> emissions, also called atmospheric carbon removal. Thus, the model scenarios typically assume fossil emissions will be twice the budget, around 1600 Gt CO<sub>2</sub>, see Fig. 8, while we leave our descendants with the expensive task of removing around 800 Gt of CO<sub>2</sub>, i.e. around 100 tonne/capita.

These model scenarios were conceived before the Paris agreement and form a basis for the ideas on what is needed to meet the climate goal. However, these modelling scenarios, are based on optimization of costs and tend to favour taking necessary costs in a distant future instead of now. Consequently, the costs for addressing the climate issue are sent to our descendants. There is a growing concern about the immoral aspect of emitting CO<sub>2</sub> that our descendants will need to remove from the atmosphere. The obvious way to make better is to reduce fossil emissions more rapidly in combination with a more rapid introduction of negative emissions at large scale.

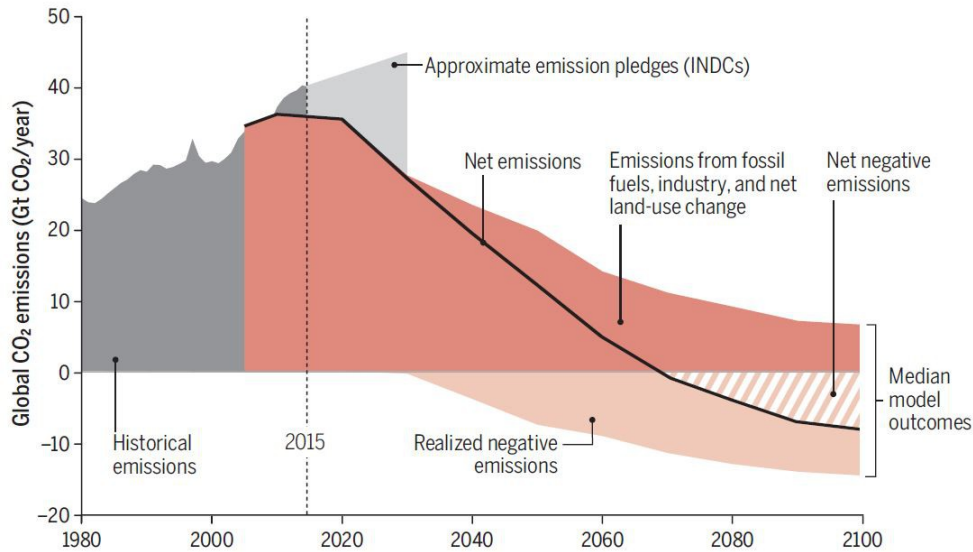


Figure 9. A median IPCC emission scenario for meeting the 2°C target. From [280]

With an assumed deficit in the carbon budget of e.g. 800 Gt CO<sub>2</sub>, and an assumed cost of negative emissions of 100 €/t the debt we leave to children and grandchildren corresponds to 10,000 € for every now living human being. Will future political leaders be able to agree on how to share these costs between countries and have their tax-payers pay their share of this debt, even if the money for practical reasons would need to be used in another country? Options for financing negative emissions are discussed in,[281].

A very attractive solution would be a producer liability for emissions of CO<sub>2</sub>, i.e. that the emitter of CO<sub>2</sub> pays for the subsequent removal from the atmosphere. This would both create the necessary incentives for rapid CO<sub>2</sub> reductions as well as the necessary funding for removing the CO<sub>2</sub> emitted.

In this context, chemical-looping of biomass could become important much earlier than expected. The necessity of negative emissions may also contribute to a more positive public perception of BECCS, i.e. Bio-Energy CCS, as compared to CCS, which has experienced difficulties in some European countries.

## 4.2 Biomass in chemical-looping combustion

In the case of biomass, CLC may also come with important advantages. Low or eliminated  $\text{NO}_x$  emissions, is one, *cf. section 3.2.4.* Another is associated with the important difficulties imposed by the aggressive alkali ash components in biomass fuels. If the alkali could be avoided in the air reactor, this could give significant reductions of maintenance costs as well as higher efficiency, see Fig. 9. It could even open up for the use of biomass fuels of high potassium content, e.g. grasses, normally avoided in boilers. However, the fate of alkali in CLC is largely unknown and further research is needed.

Some ash components may cause bed particles to become sticky and agglomerate, making the bed material impossible to fluidize, i.e. defluidization. Zevenhoven et al. studied defluidization in a lab reactor with salts to simulate biomass ash, [282]. They concluded that  $\text{KCl}$  will vaporize in the fuel reactor. Consequently, alkali chloride should not appear in air reactor. They also found that  $\text{K}_2\text{CO}_3$  will react with ilmenite and form potassium titanate. Hence, some alkali can be carried to the air reactor from potassium containing fuels if ilmenite is used as bed material. Feeding potassium dihydrogen phosphate to their test reactor Zevenhoven et al. observed bed agglomeration. This result indicates that phosphorous-containing fuels, such as agricultural residues could give difficulties.

Alkali and especially alkali chlorides is one reason that the steam parameters in solid fuel fired biomass boilers typically are lower than those used in coal fired ones. Although stem wood typically contains low amounts of chlorine, in most cases this is a too expensive fuel. Less valuable, but more demanding fractions are usually more attractive. In Bubbling Fluidized-Bed Combustion (BFBC), a technology often applied in biomass combustion, the maximum steam temperature used is typically  $550\text{ }^\circ\text{C}$ . Most of the heat in biomass CLC is produced in the air reactor. The potential for increasing the steam parameters compared to BFBC, and thus the efficiency of the Rankine cycle, seems promising. Until now, investigations on corrosion risk in CLC are scarce.

Eriksson et al. have assessed the corrosion risk in biomass-CLC using laboratory experiments,[283]. The steels chosen for testing air reactor like conditions were TP347, TP310, Sanicro28, and the high alloyed steels HRN11 and Inconel617. In these tests, material temperatures up to  $700\text{ }^\circ\text{C}$  were investigated. The steels were either exposed directly to dry air or were covered with a layer of ilmenite or ilmenite with potassium titanate. The results revealed that from a “fire-side” corrosion point of view, these material all potentially could be used in heat transfer surfaces with a much higher material temperature than in e.g. BFBC. Despite the promising results in laboratory corrosion testing at high temperature investigating

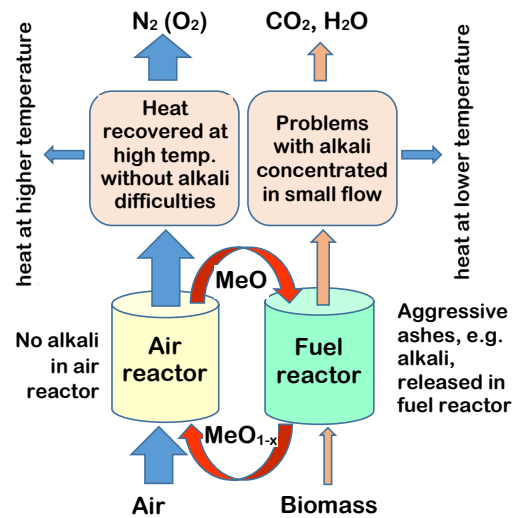


Figure 9. Advantages with respect to alkali.

super heater materials at conditions anticipated in the air reactor, it is important to bear in mind that also the strength of the material at these conditions must be taken into account.

In the case of the fuel reactor, there are no cooling surfaces and the first cooling surfaces would be downstream of the oxy-polishing step. Here the conditions would be significantly different and further research is needed. However, the heat duty of this stream is significantly smaller and cooling surfaces could be adapted to higher alkali gas content both considering materials used and cooling temperatures.

A first study on the alkali streams in chemical-looping combustion using mostly ilmenite as bed material, found that essentially all alkali was contained in the bed material. This was verified both by a mass balance showing the accumulation of alkali in the solids as well as measurements of alkali in exiting gas streams suggesting these contained only 1-2% of incoming alkali,[138].

There are a large number of existing fluidized bed boilers operating on biomass. As such, bio-CLC may be a very relevant candidate for early up-scaling of the technology, following the already mentioned up-scaling route based on using an existing CFB.

## **5 Discussion and conclusions**

The paper shows that the basis of knowledge for this novel technology is rapidly expanding. The two main barriers for the implementation of solid fuel CLC are firstly the lacking incentives for CO<sub>2</sub> capture and secondly investment risks associated with building full-scale plants with novel technology. However, there should be good hope that both these barriers can be overcome as the need for both rapidly reduced emissions, as well as negative emissions, eventually will be translated into legal and fiscal actions. The bottom-line is the much lower cost of the technology, which may be further augmented by some important advantages associated with the very clean exhaust gas that can be produced by the air reactor.

The following conclusions can be made:

- ❶ With more than 10 000 h of operational experience involving different oxygen carrier materials, fuels and reactor design, it is clear that the concept works and is possible to scale up to full scale.
- ❷ Operational experiences have advanced greatly in the last years, especially with more complex operation involving solid fuels and oxygen carriers of more complex composition.
- ❸ The large experience with a number of different oxygen carriers creates a portfolio of materials with a variation in properties and costs that forms a solid basis for the commercialization and optimization of the process. The possibility to have a choice of materials will facilitate upscaling and investment decisions.
- ❹ At least with solid fuels, and compared to conventional CFB combustion, it is clear that the added costs of chemical-looping are small, and much smaller than for competing technologies.

- ❏ The process has a number of potential advantages, in addition to inherent CO<sub>2</sub> capture, which could reduce costs further. It can also be speculated whether in some specific cases, these advantages could be great enough to motivate the CLC process even without CO<sub>2</sub> capture.
- ❏ While most CLC research has focused on fossil fuels, an important future use could be with biomass, in order to accomplish negative CO<sub>2</sub> emissions, i.e. remove atmospheric carbon.
- ❏ Although performance in full scale cannot be safely predicted, it is highly unlikely that large-scale performance would be so poor that it jeopardized the unique and principal advantage of a CO<sub>2</sub> capture technology where gas separation is inherent in the process.
- ❏ The up-scaling presents a very large barrier for the commercialization of CLC, but there could be ways to reduce the actual and perceived financial risks with building a large-scale boiler using a completely novel combustion principle.
- ❏ At present there is no market for CLC, because of lacking or insufficient incentives, but this could change rapidly. There is a clear understanding that the only way to meet the Paris agreement is to introduce strong measures directed against CO<sub>2</sub> emissions.

Thus, it can only be concluded that the future prospects for CLC should be excellent.

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