Chemical Looping Combustion of wood pellets in a 150 kW_{th} CLC reactor

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Abstract – Carbon-negative solutions have got increased attention in the recent years as being a necessary measure to mitigate climate change and limit the global temperature rise to 2°C. Bio-Energy Carbon Capture and Storage (BECCS) is at present the most mature and the only large-scale technology that can achieve CO_2 negative emissions. One attractive option for realising BECCS is by using chemical looping combustion (CLC) technology in combination with sustainable biomass as fuel (bio-CLC). Bio-CLC is a very promising CO_2 capture technology because of the potentially low energy penalty and low CO_2 capture cost. Bio-CLC pilot scale tests in a 150 kW_{th} CLC reactor system has been carried out using ilmenite as oxygen carrier and whole wood pellets as fuel. The work is part of the project "Negative CO_2 Emissions with Chemical Looping Combustion of Biomass", one of three ongoing flagship projects funded by the Nordic Energy Research. Fuel feeding rate was kept stable at a fuel power equivalent to 140 kW_{th}. The operation of the reactor was then nearly auto-thermal, as the only additional heating of the reactor was preheating of the primary air for the air reactor. The minimum oxygen demand was calculated to about 23% and the CO_2 capture efficiency varied between 94 – 97%. The specific fuel reactor inventory during the test was 140 – 180 kg/MW. This is low compared to what is used in most other studies and is mainly a consequence of the fuel reactor being a CFB type of reactor, operating close to a fast fluidization mode. Even though significant amount of additional oxygen is needed for full fuel conversion, the results may be considered good bearing in mind the relatively small size, and thus short residence time, of the reactor compared to an industrial scale reactor, and the low-cost oxygen carrier material used.

1 Introduction

In order to limit the global temperature rise to the internationally adopted goal of 2°C [1], CO_2 capture and storage (CCS) will be one of several technologies needed. In the 2°C scenario, CCS will contribute with about 12 – 15% of the needed CO_2 emissions reduction through 2050 [2]. In the recent years, carbon-negative solutions such as BECCS (Bio-Energy Carbon Capture and Storage), have got increased attention and have also been highlighted as necessary to meet the 2°C limit [3]. This is further strengthened through the study of Kriegler et al. [4] arguing that versatile technologies such as CCS and bioenergy are most important for climate change

mitigation since their combination will produce significant negative emissions. BECCS is the most mature and the only large-scale technology that can achieve CO_2 negative emissions. It has a wide range of applications, such as in different biofuel production technologies, biomass-fired combined heat and power plants and in co-fired biomass/coal plants ([3], [5]).

One attractive and promising option for realising BECCS is by using chemical looping combustion (CLC) technology. The development of the CLC technology has been moving rapidly forward during the last 15 years. The process is proven in several lab scale and pilot scale setups around the world [6] and successful operation has been obtained for durations of up to several weeks. CLC is a very promising CO_2 capture technology because of the potentially low energy penalty and low CO_2 capture cost [7]. The process can be described as a special type of an oxy-fuel process, but with inherent air separation where a metal oxide is alternately oxidised and reduced, taking up oxygen from the air in an air reactor and supplying the oxygen to the fuel in a fuel reactor. The principle is illustrated in Figure 1. The air and fuel are not mixed and the exhaust gases from combustion contain almost only CO_2 and H_2O so that CO_2 separation can be performed simply by condensation of the H_2O. Much work has been conducted on development of the metal oxide materials, the so-called oxygen carriers (OC), as it is critical to develop materials with high oxygen transport capacity, high durability for a long lifetime, being environmentally viable and preferably have low production cost. The oxygen carrier materials have mainly been based on oxides of Fe, Mn, Cu, Ni and Co, either synthetically made or from natural ores such as e.g. ilmenite.

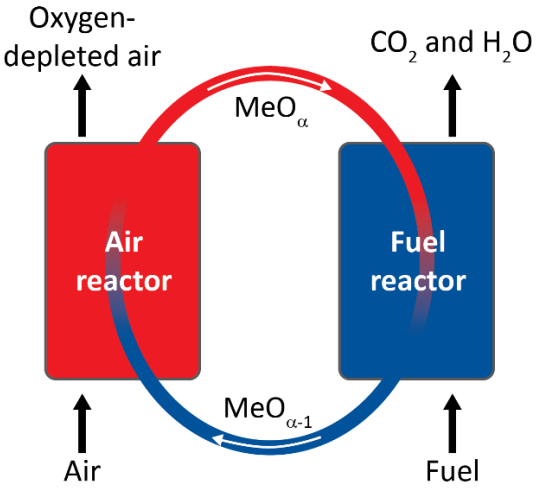

Figure 1: Principle of CLC.

CLC of solid fuels has been reported from 19 pilot plants, with a fuel input power ranging from 0.5 kW_{th} to 4 MW_{th} and with more than 2700 hours of operation [8]. The most common configuration is two interconnected fluidized bed reactors, where the air reactor is arranged as a fast velocity riser and most of the fuel reactors are of the bubbling bed type. Traditional fluidized bed technology was to a large degree developed for conversion of solid fuels, being fuel flexible, giving high combustion efficiency and low emissions of NOx. A CLC plant for solid fuels can be obtained by extending this to a two-reactor system and using oxygen carrier particles as the bed material. It will then have the advantage of sharing significant similarities with commercially viable fluidized bed technology [7].

CLC of solid fuels has mainly been done using coal as fuel, however, biomass has more recently become an attractive option for CLC because of the negative CO_2 emissions that can be obtained ([8], [9]). The Nordic countries are in a very good position for developing and deploying such a Bio-CLC technology [10]. They have huge forests and possibly large supplies of sustainable biomass. Finland, Sweden and Denmark are among the world-leading countries with respect to heat and power generation from biomass, including manufacturers of fluidized bed technology. Norway has a similar position within CCS, especially through the on-going Norwegian full-scale CCS project involving CO_2 capture from potentially three industrial sites, CO_2 transport by ship and an offshore CO_2 storage facility. The storage facility will have large capacity and possibility for other capture projects to join to get better assurance of CO_2 supply and lower storage cost per tonne ([11] - [13]).

This paper presents results obtained in the 150 kW_{th} CLC reactor system at SINTEF Energy Research, using ilmenite as oxygen carrier and whole wood pellets as fuel. The work is part of the project "Negative CO_2 Emissions with Chemical Looping Combustion of Biomass", one of three ongoing flagship projects funded by the Nordic Energy Research [10].

2 Experimental setup

2.1 Overview of the CLC pilot plant

The CLC reactor system consists of two circulating fluidized bed reactors as shown in Figure 2. The two reactors, the air reactor (AR) and fuel reactor (FR) respectively, are interconnected through particle loop seals that works as gas locks to ensure that only the oxygen carrier particles are transferred between the reactors. In addition, particles are also transferred from the fuel reactor to the air reactor through the lifter, which is fed from the bottom of the fuel reactor.

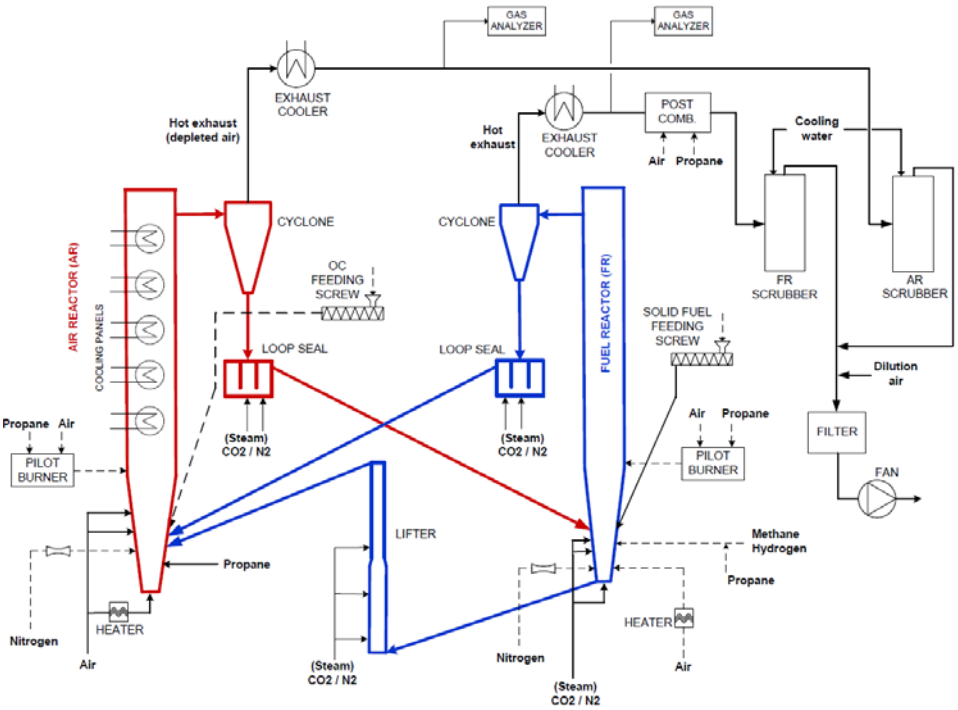

Figure 2: Reactor system.

Both reactors are 6 m tall of which the first 1 m is a conical bottom section. The remaining 5 m cylindrical sections have internal diameters of 230 mm (AR) and 154 mm (FR). The system has one solid fuel feeding screw and one oxygen carrier feeding screw which makes it possible to refill particles during operation. The reactors are heated up by hot air and fuel that are introduced into the particle beds. Pilot burners are mounted above the bed to ensure safe ignition of the injected fuel. In addition, a 10 kW electric heating wire is mounted on the bottom part of the fuel reactor. The heating wire will be in operation during the heat-up sequence and if the FR temperature gets too low. During CLC operation, the reactor temperature is controlled by adjusting the primary air preheat temperature to the air reactor. If this is not enough, it is possible to inject propane into the air reactor, however, this is generally not necessary and in the present test it was only used during the heat-up sequence. In addition, five cooling panels are mounted within the air reactor if cooling is needed. The system is originally designed for operation on methane as fuel gas at a maximum fuel power of 150 kW [14].

The system has two Teledyne 7500 IR gas analyzers measuring the CO, CO_2 and CH_4 concentration in the exhaust from each of the two reactors. In addition, a two-channel Servomex 4900 for O_2 measurements from each reactor, as well as a Horiba PG250 gas analyzer for extended CO_2 range was connected during the test. N_2 was used as fluidization gas for the loop seals, fuel reactor and lifter. The system is designed to use steam as fluidization gas, but there was no steam boiler available during the present test.

2.2 Test conditions

The following results are gathered from a one-day test using ilmenite as oxygen carrier and Arbaflame black wood pellets as fuel. This was the first test using solid fuels in the present CLC reactor system. The ilmenite particles, which were provided by Titania AS, had a particle size (d_{50}) of 80 μm and a bulk density of about 2600 kg/m^3. The wood pellets (diameter 8 mm, typical length 30 mm) are produced according to the special process of Arbaflame, which results in slightly different properties compared to common types of wood pellets. According to an analysis of these pellets performed by VTT in Finland, the volatile content is 79.1% and the fixed carbon content is 20.4% on dry basis. The moisture content was 4.9% and the lower heating value was 19.3 MJ/kg.

Figure 3 shows the reactor temperatures, air preheat temperature, wood pellets feeding rate and additional propane firing in the air reactor from the end of the heat-up sequence and throughout the test. During the reactor heat-up, combustion took place in both air and fuel reactor as propane and air was fed to both reactors. At about 15:30, feeding of wood pellets was started and the addition of propane was reduced and stopped at about 15:40. From there on, the reactor operated as a chemical looping reactor with wood pellets as the only energy supply, except for the electric preheat of the primary air to the air reactor. The first two hours of CLC operation was used to tune in the process and solve practical issues with gas analysers etc. Unfortunately, the system shut down at about 18:40 because an over-temperature switch in the AR triggered the emergency shutdown system. After recovering from the shutdown, the fuel was changed from whole to grinded wood pellets. During this period, the fuel reactor pressure was very fluctuating, and occasional clogging of the cyclones took place. This can be ascribed to the fuel feeding system, as very uneven feeding was observed during the feeding tests when using

grinded pellets compared with using whole pellets. The test was finished with a period of feeding whole pellet again. The best results achieved from the test was obtained in a 40-minute period from 18:00 until the emergency shutdown at 18:40. The rest of the results presented below are therefore from this period. As can be seen from Figure 3, in this period there was a stable fuel feeding of about 26 kg/h, equivalent to 140 kW based on the lower heating value. Air reactor temperature was about 1000°C and fuel reactor temperature in the range 960 – 980°C. The primary air preheat was around 820°C whereas the additional propane feeding to the air reactor was zero.

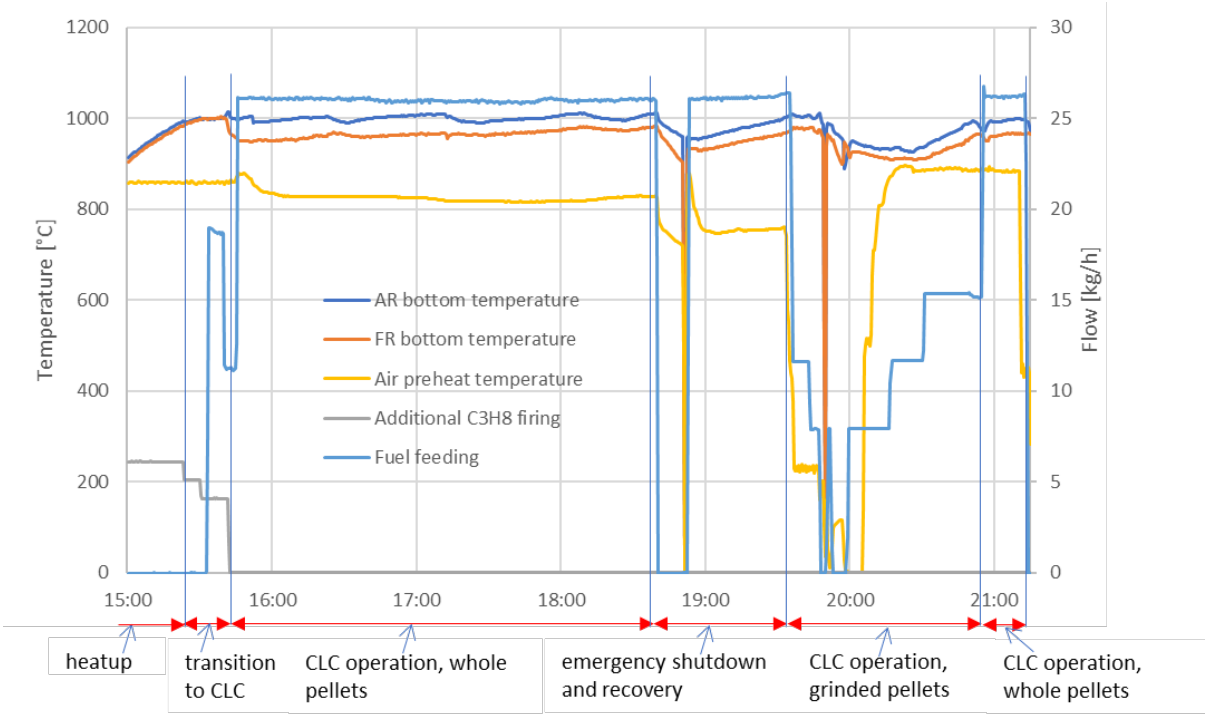

Figure 3: Temperatures and fuel feeding during test.

3 Results and discussion

Figure 4 shows the gas concentrations at the AR and FR outlets. Both the fuel feeding (26 kg/h) and gas flows was kept constant during the period, which also resulted in quite stable emissions. A slight decrease in FR CO and CH_4 concentration with a corresponding slight increase in CO_2 concentration (about 42 – 44%) can be observed during the period. This can be explained by an increased FR inventory as oxygen carrier particles were fed into the reactor leading to higher oxygen transport and fuel conversion. The rest of the gases out of the FR is mainly N_2 which is used for fluidization, and H_2 which was not measured.

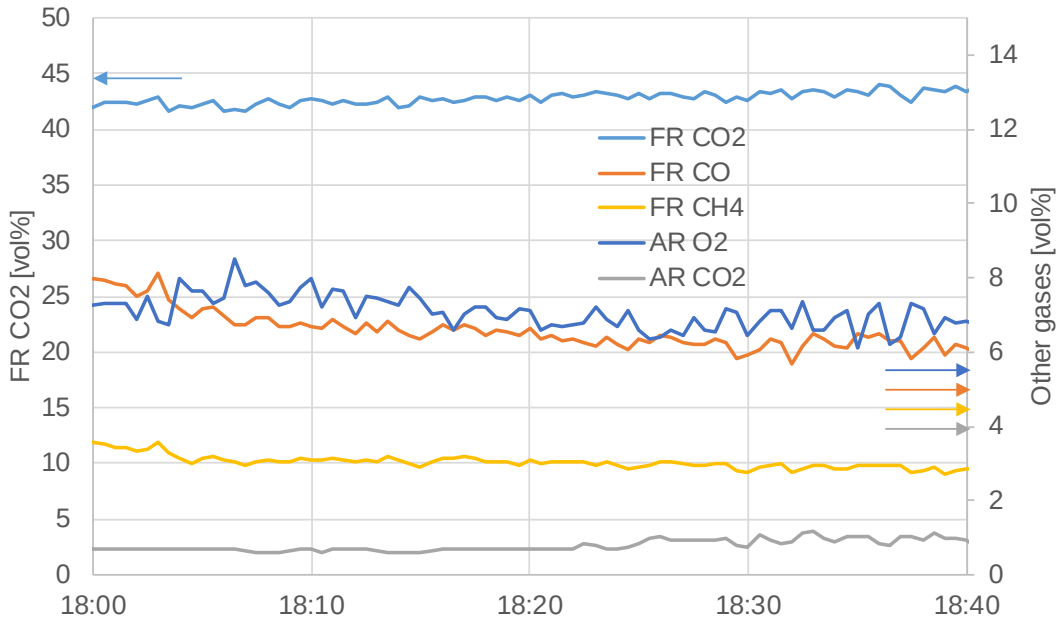

Figure 4: Reactor outlet gas concentrations.

A slight decreasing trend can also be seen for the O_2 concentration from the AR, as more oxygen is taken up from the air when the oxygen carrier inventory increases. The CO_2 concentration from the AR is caused by char residue following the OC particles from the FR and combusting in the AR. The amount of CO_2 produced in the AR is not negligible, as it results in a reduction in the CO_2 capture efficiency to about 95%. However, a certain "leakage" of char to the AR must be expected, as no carbon stripper is mounted, and the FR operates in the turbulent to fast fluidization regime. This makes it very likely that some char particles are entrained in the upward flow in the reactor and partly follows the OC particles to the AR and partly follows the exhaust gas out of the cyclone and ends up in the exhaust filter. Some char particles were observed in the exhaust system but the amount was not quantified since the system was not yet made ready for that. However, compared to the amount of fuel introduced during the test, the amount can be considered low.

Figure 5 shows the CO_2 capture efficiency, the CO_2 yield and the O_2 demand calculated in two different way. The O_2 demand, which can be defined as the additional oxygen needed to combust all unburnt species, relative to the stoichiometric amount of oxygen, can simply be calculated from the oxygen balance of the AR and the chemical composition and mass flow of fuel. It can also be calculated from the gas analysis of the FR exhaust. However, the hydrogen concentration is part of the calculation, and this was not measured during the test. The hydrogen comes only from the chemical bonded hydrogen and the moisture in the fuel, as no steam was added in the test. The green line represents the O_2 demand if one assumes a fixed H_2/CO molar ratio of 1.3. The line roughly coincides with the yellow line based on the AR O_2 balance and the fuel consumption, which suggests that the H_2 level was in this range. Others, e.g. Mendiara et al. [15], has found that the H_2 level is similar or higher than the CO level. Tests performed by Pikkarainen et al. [16] on ilmenite and black wood pellets also indicates that the H2/CO molar ratio is in this range. As shown in Figure 5, the O_2 demand during the relevant test period was in the range 23 – 28 %. This is in the same range as what was obtained in [15] using pine sawdust as fuel and the iron-based Tierga ore as oxygen carrier. It

is also in the same range as found by Linderholm et al. [17] who were using the same type of black wood pellet fuel but a sintered manganese ore as oxygen carrier. The obtained O_2 demand is somewhat lower than what was found in [16].

The CO_2 capture efficiency is calculated using the molar flow of CO_2 escaping out through the air reactor and the molar flow of carbon fed to the fuel reactor via the wood pellet fuel. The CO_2 out from the air reactor results from burn-out of char coming from the fuel reactor. Char losses through the FR exhaust is not included in the calculation, however, in an industrial CLC unit it would be expected that unburnt compounds in the FR exhaust will be oxidized in the oxygen polishing step just downstream of the fuel reactor [15]. The calculated capture efficiency was in the range 94 – 97 %. This is slightly higher than the values found in [16] which was about 90%, but lower than found in [15] and [17] which both obtained values very close to 100%. The CO_2 yield is a measure of the conversion efficiency of carbonaceous gases into CO_2 in the fuel reactor. The obtained values range from 79 – 83 %.

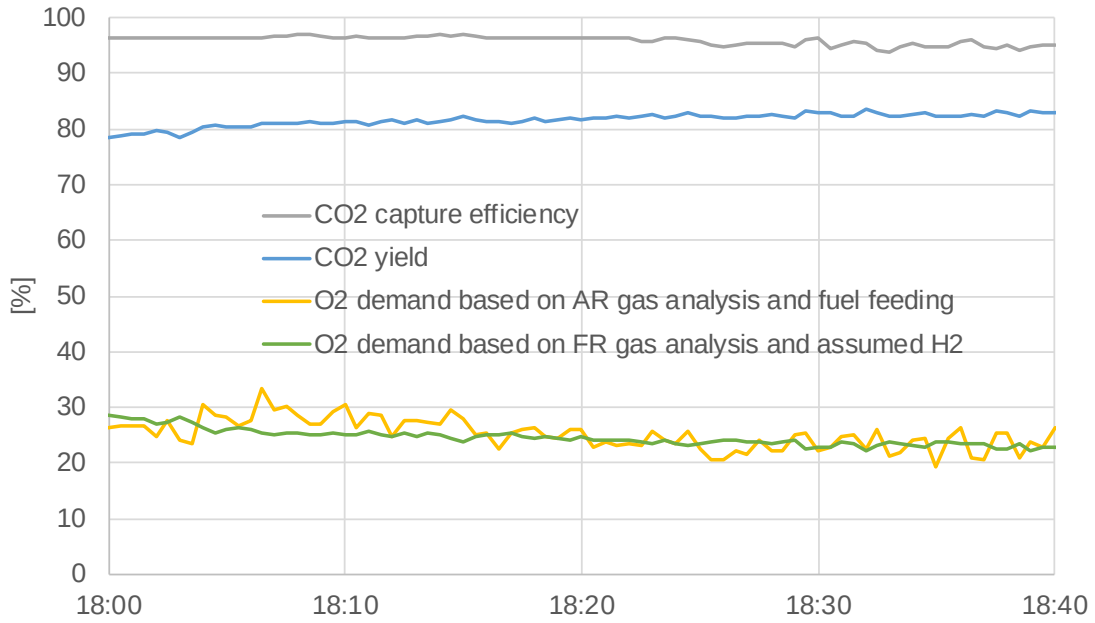

Figure 5: Efficiencies.

When comparing the results with the other studies, some notable differences should be mentioned. The fuel power in the present study was about 140 kW, whereas it was 0.5 kW in [15], 15 – 22 kW in [16] and 67 kW in [17]. The other studies used steam as fluidising gas, which may increase the gasification rate of the char which is a critical factor to fully convert the fuel before it leaves the fuel reactor. The fuel reactor temperatures were in the same range except for [16] where the temperature was about 100°C lower than in the present study due to a main challenge in achieving the desired high temperature when operating on ilmenite.

Figure 6 shows the oxygen carrier inventories in the AR and FR, and the specific FR inventory. The inventories are calculated from the pressure recordings and geometry of the reactors. During the period, about 25 kg of oxygen carrier was fed into the reactor system, which can clearly be seen as an increasing trend on the inventories. The geometry of the reactors and the relatively high velocities results in low reactor inventories and residence

times compared to reactors using larger area bottom parts where the fluidization is more towards the bubbling regime. The advantage of the current design is that particle concentration is high in the whole reactor, resulting in good mixing of particles and fuel. The disadvantage is that the gasification of the char residue is limited by the low residence time.

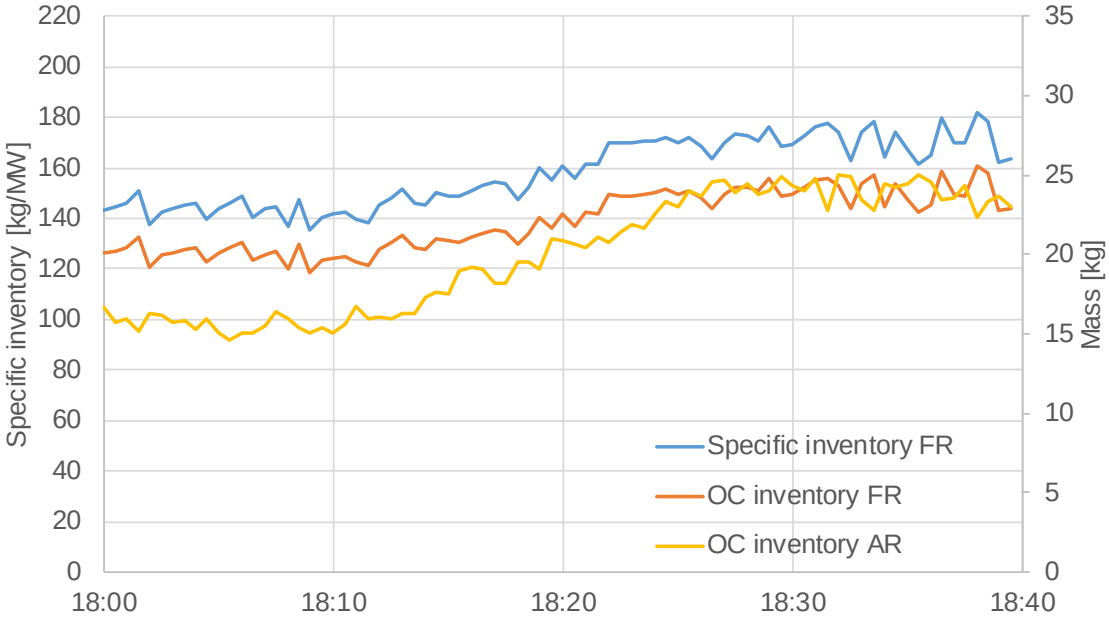

Figure 6: Oxygen carrier inventory.

Figure 7 shows the riser mass flow and the superficial velocities in the reactors. The riser mass flow is not a measure of the particle circulation, but is the calculation of particle mass flowing upwards in the upper half of the reactor. The calculation is based on the pressure difference between the upper and middle part of the reactor, the superficial velocity and the terminal velocity of the particles.

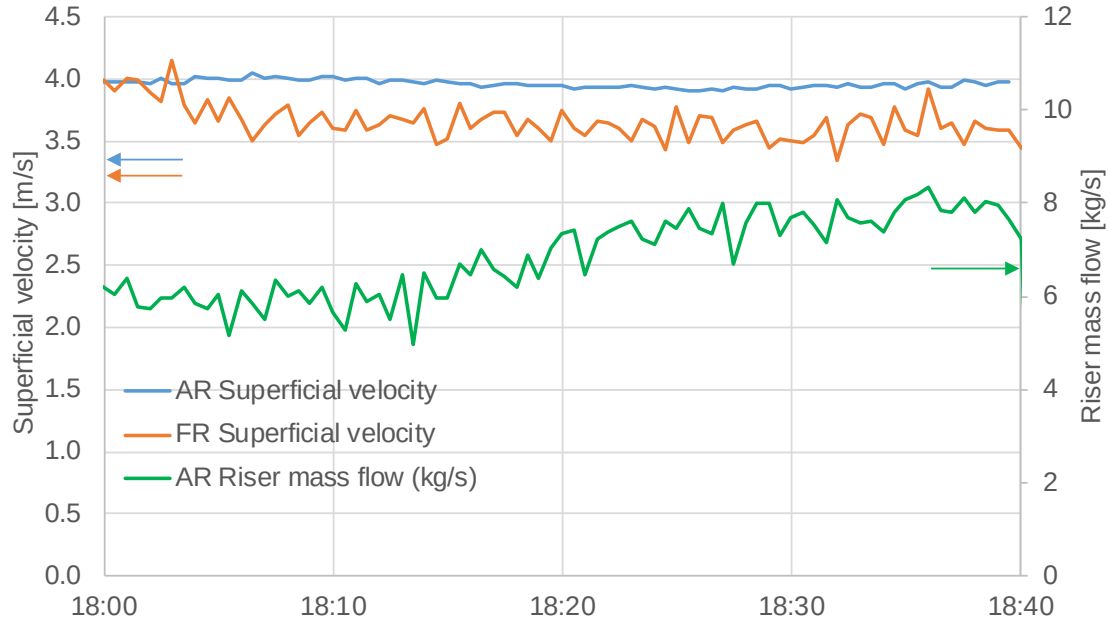

Figure 7: Reactor superficial velocity and riser mass flow.

Because the particles tend to flow upwards in the central part of the riser and fall downwards close to the walls, the riser mass flow is larger than the real net mass flow out of the reactor. It is possible to find a correlation between the riser mass flow and the global circulation rate e.g. by using the method of Linderholm et al. [18].

4 Conclusions

Chemical looping combustion tests using wood pellets as fuel and ilmenite ore particles as oxygen carrier has been performed in the 150 kW CLC reactor at SINTEF Energy Research. Both whole 8 mm pellets and crushed /grinded pellets where tested, but only the whole pellets resulted in stable feeding and stable reactor conditions. The present results are from a period of the test where the fuel feeding rate was kept stable at 26 kg/h, equivalent to 140 kW based on lower heating value. This is a rather high fuel feed value when compared to other studies found in the literature. The operation of the reactor was then nearly autothermal, as the only additional heating of the reactor was preheating of the primary air for the air reactor. The minimum oxygen demand was calculated to about 23% and the CO_2 capture efficiency varied between 94 – 97%. The loss of char residue from the fuel reactor to the exhaust was observed but not quantified. It was considered low when considering the amount of fuel introduced during the test. The specific fuel reactor inventory during the test was 140 – 180 kg/MW. This is very low compared to what is used in most other studies and is mainly a consequence of the fuel reactor being a CFB type of reactor, operating close to a fast fluidization mode. Even though significant amount of additional oxygen is needed for full fuel conversion, the results may be considered good bearing in mind the relatively small size, and thus short residence time, of the reactor compared to an industrial scale reactor, and the low-cost oxygen carrier material used.

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