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Industrial steam generation with 100% carbon capture and insignificant efficiency penalty – scale-up of oxygen carrier for chemical-looping combustion using environmentally sustainable materials

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Deliverable D4.5

Report on performance and attrition testing on 25 kg batch of impregnated material

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Chalmers	**DELIVERABLE 4.5**

Report on performance and attrition testing on 25 kg batch of impregnated material

SUCCESS - Industrial steam generation with 100% carbon capture and insignificant efficiency penalty - scale-up of oxygen carrier for chemical-looping combustion using Environmentally sustainable materials	*Keywords:* *Fe_2O_3, Al_2O_3, impregnated, continuous operation, attrition*
Work package: 4 – Attrition testing and performance verification of material	
Involved partners: Chalmers, CSIC, JM **Authors**: Tobias Mattisson, Peter Hallberg	*Dissemination level: PU* *Nature: R*

Objective

The objective of work package 4 is to evaluate the material from work packages 1, 2 and 3. The material will be evaluated both with respect to attrition as well as reactivity.

Summary

In task 2.4 a 25 kg batch of impregnated oxygen carrier material was produced by JM. The material chosen for the 25 kg batch was iron oxide based, with a composition of 20 wt% Fe_2O_3 on γ-Al_2O_3 from Sasol (Puralox SCCa-150/200). The material underwent testing at Chalmers, using a continuous CLC unit designed for 10 kW_{th}. The unit operates at velocities which are expected in an industrial unit, and thus it is believed that attrition levels could correspond rather well with those in real units. Tests were performed for 40 h operation, with 30 h of these with natural gas. Fuel conversion was monitored for a range of different operational parameters. The loss of fines was also monitored during the operation giving an indication of particle lifetime. The fuel conversion ranged from 60 to 85 % and increase with specific solids inventory as well as temperature. However, the specific solids inventory was very high, 1650 kg/MW for the case where best conversion was found. The attrition measured as material < 45 µm produced was 0.1 % of total inventory per hour of hot fluidization.

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1. Introduction

In WP2, there is a development of oxygen carrier based on impregnation of viable oxygen carriers with low attrition and high resistance towards sulphur. There has been a focus on iron-based materials, impregnated on alumina supports. A large number of these type of materials have been evaluated in this workpackage, see D2.1 and D2.2. The material selected for scale-up to 25 kg was Fe_2O_3 impregnated on γ-Al_2O_3 from Sasol (Puralox SCCa-150/200). The nominal composition was 20 wt% Fe_2O_3 on γ- Al_2O_3. A batch of 25 kg of material was produced by JM and dispatched to Chalmers for testing with natural gas using Chalmers 10 kW continuous unit. This gas-fired reactor operates using velocities in the air reactor and riser corresponding to those which are expected in a commercial circulating fluidized bed boiler or CFBB. The velocities in the riser are between 3-4 m/s, but the velocities in the cyclone inlet considerably higher. This means that realistic attrition rates are expected utilizing this reactor unit.

2. Experimental

2.1 Material

The oxygen carrier particles were prepared by JM at their facility in Billingham, UK. The selected material was Fe_2O_3 on an Al_2O_3 support, more specifically Fe_2O_3 impregnated on γ-Al_2O_3 from Sasol (Puralox SCCa-150/200). An iron nitrate solution was prepared and used as the precursor of the active iron oxide. The alumina particles were impregnated in two steps using the nitrate solution, followed by drying and calcination at 950°C. A more detailed description will be given in Deliverable 2.4. The materials tapped bulk density was 1090 kg/m^3 which was considerably lower than other materials tested in the same unit previously. There was a large fraction of particles below 125 µm, as can be seen from figure 1. The low density combined with the relatively high fraction of small particles indicated that these particles might not be suitable for this reactor. This is due to the rather high rate of elutriation of particles seen previously from the air reactor, likely due to an unoptimized cyclone at the exit of the riser.

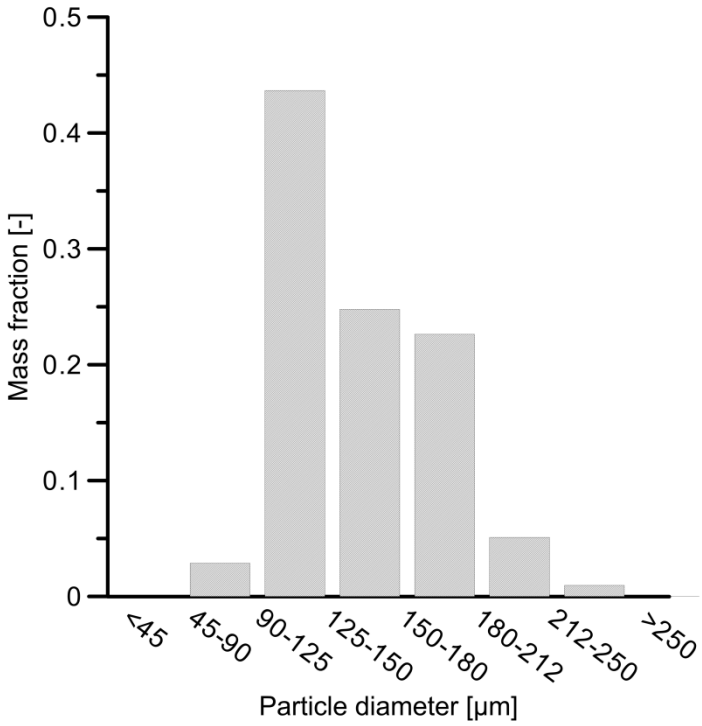

Figure 1. Fresh particle size distribution for the selected impregnated particles

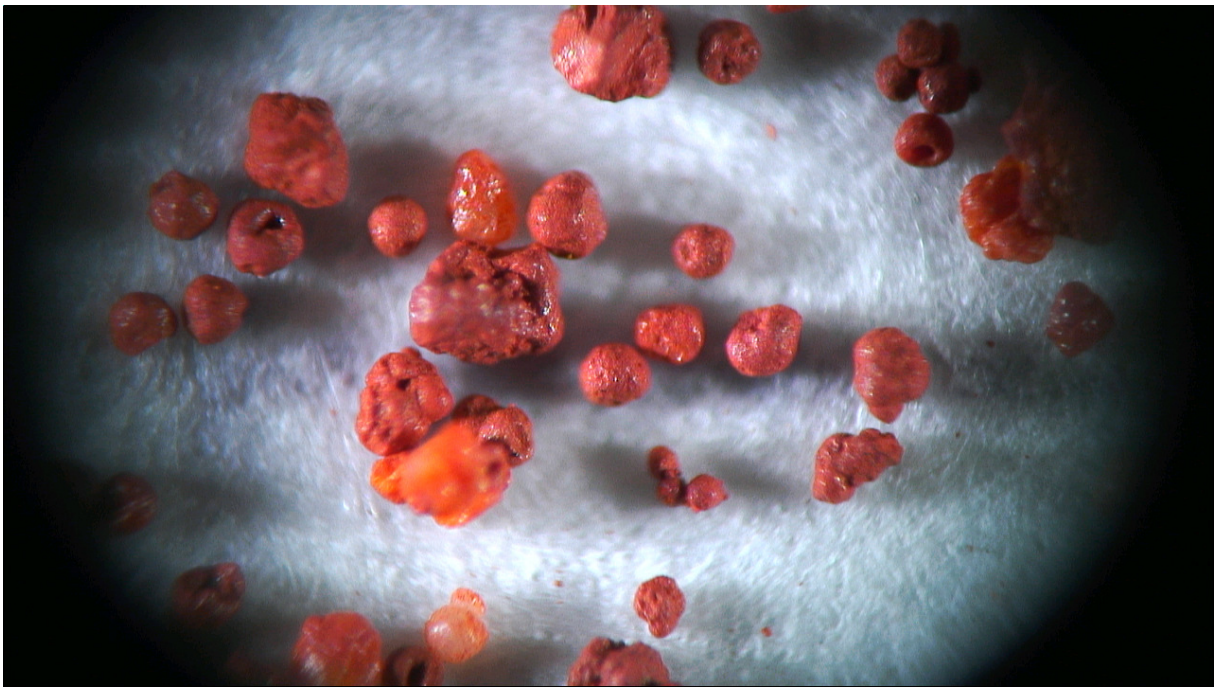

Figure 2. Light microscope image of the fresh particles

2.2 Continuous testing

The reactor system (*figure 3*) used was built in 2002-2003 in the project GRACE. The first successful demonstration of continuous CLC was performed in this unit. Few changes have

Table 1. Operational conditions for tests.

Parameter	Value
Fuel flow	5-9 L_n/min (3.2-5.8 kW_{th})
Fuel reactor temperature	900 – 970°C
Bed mass in fuel reactor	≈ 3,3 kg
Total bed mass	9-12 kg
Air reactor flow	160-180 L_n/min
Nitrogen flow in loop-seals (each)	4 L_n/min
Total time fluidized under hot conditions	40 h
Total time with natural gas	30.3 h

been made to the system since then. The nominal effect is 10 kW_{th} with natural gas. An air reactor (∅ = 15 cm) that thins out to a riser (∅ = 8 cm) drives the global circulation of solids. After the riser, gas is separated from the particles in a cyclone. From the cyclone the particles fall down into a particle loop seal that prevents gas leakage between the air- and fuel-side. Below the loop seal particles continue down to one side of a bubbling fuel reactor. The fuel reactor is shown in Fig. 4. On the other side of the FR an overflow exits leads the particles, via the next loop seal, back to the air reactor. A vertical plate in the middle of the FR prevents particles from bypassing the bed.

The gas that leaves the cyclone and FR is first passively cooled through finned pipes before sample streams are withdrawn and led to a gas conditioning system. After the gas conditioning system, where the gas is cooled and filtered, CO, CO_2, CH_4 and O_2 are measured continuously. There is also a possibility to analyse gas in a gas chromatograph at intervals to check N_2 and H_2. The gas from the cyclone is then led to a bag filter that separates the elutriated particles before it goes to the chimney. The gas from the FR goes through a water seal with the dual purpose of collecting condensed steam and controlling the pressure in the FR.

Table 1 shows the main parameters studied in this work. The power was varied between 3-6 kW. As the mass of oxygen carriers in the fuel reactor is expected to be rather constant for all tests, the specific solids inventory was different for all cases. The total time where the particles were fluidized under hot conditions with recirculation was 40 h, whereof 30 h with natural gas.

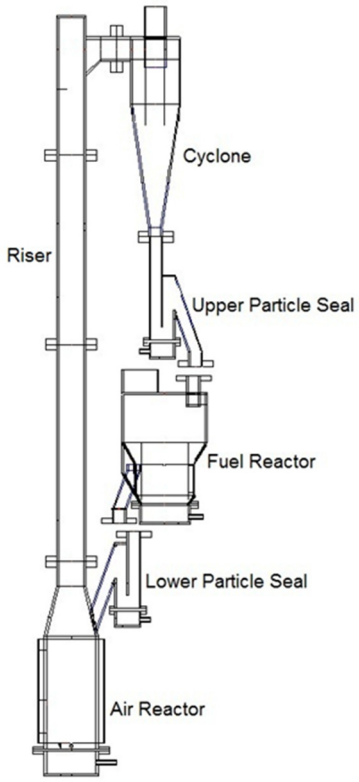

Figure 3. A schematic picture of the 10 kW fluidized-bed reactor. Auxiliary equipment not shown.

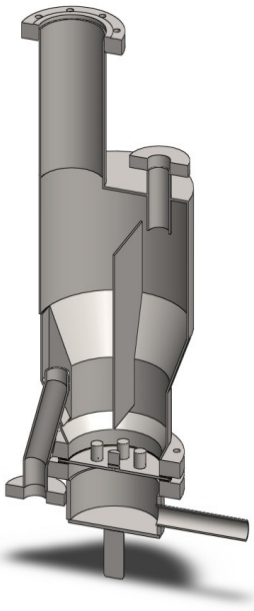

Figure 4. A 3 dimensional image of the fuel reactor with the vertical plate.

2.3 Data evaluation

The ability of an oxygen carrier to convert fuel to CO_2 is expressed by the fuel conversion γ. The calculation is made based on a carbon balance over the reactor system. Equation (1) represents CH_4 conversion.

$$\gamma_{CH4} = \frac{p_{CO_2}}{p_{CO_2} + p_{CO} + p_{CH4}} \qquad (1)$$

where p_i is the outlet partial pressure of component i. Expression (1) gives an indication of the ability of the oxygen carrier materials to convert the fuel to CO_2. This could of course be dependent upon the degree of oxygen carrier conversion, which is proportional to the degree of solids recirculation. In this unit the actual circulation was not possible to measure directly.

3. Results and discussion

3.1 Attrition

The total solid inventory was around 9 – 12 kg. As anticipated the elutriation of solids was considerable, around 0.7 kg/h. Most of the elutriated particles were found in the bag filter located after the air reactor. In order to separate the fines (< 45 µm) all elutriated material was wet sieved and subsequently dried. After sieving and drying the particles, all bigger than 45 µm were returned to the reactor. Since the elutriation was faster than the handling of the elutriated material fresh particles were used to replace the elutriated the first few times the filter was emptied. Because of this a total of 17.6 kg of particles were used although at most 12 kg were in the reactor on any occasion. Figure 5 showing the fine fraction of the elutriated material. Only including the operation with fuel the attrition is 0.14%wt/h. If circulating operation without fuel is included in this number the attrition is 0.10%wt/h.

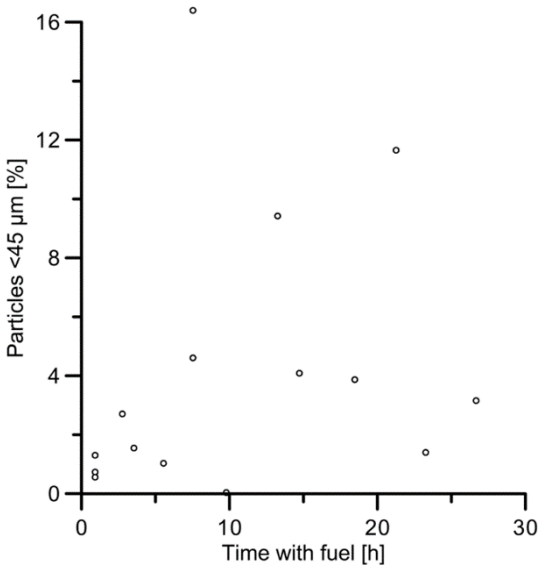

Figure 5. Fraction of fines in the elutriated material as a function of operating time.

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3.2 Fuel conversion

Many different parameters was important for fuel conversion. The influence of fuel reactor temperature and specific fuel reactor inventory is shown in figure 6. Tests were performed using specific solids inventories between 619-1650 kg/MW. For the case with 1650 kg/MW_{th} the fuel flow was not high enough to fluidize the bed and therefor nitrogen was added to the fuel to increase gas velocity. This of course, has the effect of diluting the methane stream, which could affect the reactivity. From the figure it is evident that the gas yield varied between 60-85%. The gas yield increased as a function of solids inventory and temperature. Another important parameter for fuel conversion was the total inventory of particles. Unfortunately the total inventory was impossible to keep constant due to the high elutriation of particles. The total inventory also affects the circulation of particles which is an important parameter for fuel conversion. In the current experiments the air flow was varied between 160-180 L/min, but no observable effect on conversion was seen.

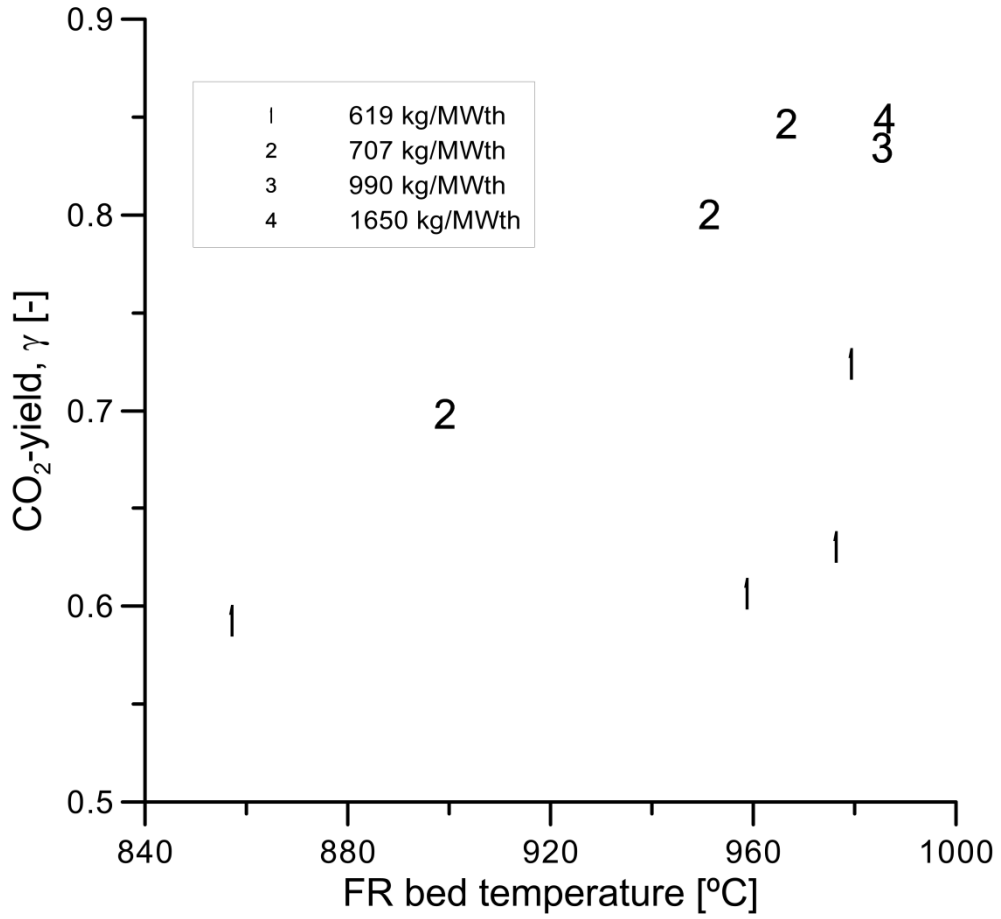

Figure 6. CO_2-yield as a function of FR bed temperature and specific fuel reactor inventory.

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4. Conclusions

This deliverable reports on the results of testing of a Fe-based oxygen carrier prepared by impregnation. The material was tested at realistic conditions for 30 h with methane and an additional 10 h under hot conditions. Despite the low density it was possible to use this material without any major problems. However it was not possible to come close to complete fuel conversion even at 980 °C and 1650 kg fuel reactor inventory per MW_{th}.