Attrition of oxygen carrier particles in fluidized bed – basic theory and screening measurements with a customized jet cup test rig

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Abstract

This article provides an overview of expected attrition phenomena during chemical-looping processes in interconnected fluidized-bed reactors, and also describes a customized method for measuring attrition resistance of oxygen-carrier particles. The proposed method is similar to the jet cup method, but has been scaled down in order to be suitable for samples as small as 5 g. Experimental results for materials, which previously have been used in continuous experiments in different reactor systems, are presented in brief. The attrition behaviour of materials during continuous operation was compared to results obtained with the jet cup test rig, and a strong correlation was established. Also, a weaker correlation between oxygen carrier performance and the commonly used crushing strength index was found. Some general guidelines with respect to particle attrition during chemical-looping combustion are provided. Materials that show good attrition resistance in the jet cup tests are much more likely to perform well in real operation. Also particles with a crushing strength greater than 2 N are more likely to perform well compared to softer particles. Composite materials with NiO or Fe_2O_3 as active phase and Al_2O_3-, $NiAl_2O_4$- or $MgAl_2O_4$-based support, as well as materials based on the $CaMnO_{3-\delta}$ perovskite structure, are among the materials that were found to have high attrition resistance, and which also improved further following operation with fuel. In contrast, combined iron-manganese oxides $(Fe_xMn_{1-x})_2O_3$, and materials containing smaller or larger amounts of either CuO or ZrO_2 experienced reduced attrition resistance during operation with fuel, and usually also had low attrition resistance to begin with.

1 Introduction

The concept of using solid oxygen carrier particles as bed material in fluidized bed reactors has many promising applications:

- Chemical-looping combustion (CLC) is an innovative method for combustion in which fuel is oxidized using two separate reactor vessels, one air reactor (AR) and one fuel reactor (FR). A solid oxygen carrier (Me_xO_y) performs the task of transporting oxygen between the two reactors. In the fuel reactor, it is reduced by the fuel, which in turn is oxidized to CO_2 and H_2O. In the air reactor, it is oxidized to its initial state with O_2 from air. The operating temperature of each reactor is in the range of 800-1000°C, and the net energy released in the reactor system as a whole is the same as in ordinary combustion. Since the fuel is not mixed with N_2 from air, condensation of the steam generated in the fuel reactor is sufficient to obtain almost pure CO_2. Hence the procedure could be considered an ideal technology to provide CO_2 for carbon capture and storage, which has been recognized as a very important option for climate mitigation. Chemical-looping combustion has been proven to work well and there is currently more than 4800 h of operational experience using circulating fluidized bed systems in sizes up to 140 kW_{th}[1].

- Chemical-looping reforming (CLR) works in a similar fashion but the air to fuel ratio is greatly reduced in order to achieve partial oxidation rather than combustion. The concept has been demonstrated in practical operation for both generation of synthesis gas ($CO+H_2$) from natural gas[2], and for cracking of tar components during biomass gasification[3].

- Chemical-looping steam reforming (CLRs) and the steam-iron reaction (SIR) are methods for generation of pure hydrogen from other fuels. The former is basically a combination of ordinary steam reforming and chemical-looping combustion[4]. The latter generates H_2 by oxidizing a reduced oxygen carrier with H_2O rather than with air, while utilizing a three reactor setup[5].

- Oxygen carrier aided combustion (OCAC) is a novel combustion concept that promises reduced emissions and improved efficiency for power generation from biomass and coal. It is similar to ordinary fluidized bed combustion, with the difference that active oxygen carrier particles are used as bed material rather than inert sand. The oxygen carrier helps to even out the oxygen potential and heat generation throughout the combustion chamber. In oxygen lean parts of the reactor, the endothermic reduction of oxygen carrier particles will occur, while the exothermic oxidation occurs in oxygen rich parts. In practice, problems associated with poor mixing between air and fuel and hot-spots are reduced. The concept has been demonstrated in large pilot scale of 12 MW_{th}[6]. Reduction of CO emissions with up to 80% and NO emissions with up to 30%, reduced fouling of heat exchanger surfaces and the possibility to decrease the overall air ratio of combustion for improved thermal efficiency have been demonstrated.

The concepts outlined above, henceforth referred to as chemical-looping processes, utilize solid oxygen carrier particles to transfer oxygen between different reactor vessels, or in some instances between different reaction zones within a single reactor. The oxygen carrier particles typically are of similar size (90-250 μm) and shape as fine sand. Commonly used oxygen carriers include metal oxides of the d-block transition metals Fe, Mn, Cu and Ni. To increase reactivity the active materials are often dispersed on inert support materials such as Al_2O_3, $MgAl_2O_4$ or ZrO_2. Most work has focused on monometallic metal oxides as the active phase, but some work has been done also on combined oxides such as for example $(Mn_zFe_{1-z})_yO_x$ and $CaMn_xTi_{1-x}O_{3-\delta}$. Oxygen carrier particles can be manufactured with different methods, of which spray drying and impregnation are among the more interesting for large-scale applications. Naturally occurring ores and waste materials from industrial activities have also been examined as oxygen carriers and are considered to be very interesting, in particular for solid fuel applications and for oxygen carrier aided combustion. More than 900 different materials have been investigated with different combinations of reactors and fuels and many have been used also in actual operation[1,7].

Chemical-looping processes are typically realized by use of fluidized-bed reactors. The arrangement is similar to ordinary circulating fluidized bed boilers (CFB). The oxygen carrier particles constitute the bed material and are continuously transported through the different parts of the reactor system. The phenomenon attrition is bound to be of uttermost importance for chemical-looping processes that utilize this configuration. This is because particles in fluidized beds are subject to high velocities that can be expected to cause attrition.

While there has been rapid progress within the research field of chemical-looping processes during the past decade, the attrition behaviour of oxygen carrier particles has received limited attention. What is clear is that many types of materials are subject to physical degradation (fragmentation, dust formation etc) after only a few hours of operation, while others remain practically unaffected over the course of hundreds of hours of operation and thousands of redox cycles. This is an important observation since severe attrition may be unacceptable in industrial applications. Because of this it is vital to gain insights about relevant attrition mechanisms, and to understand why some materials perform well while others do not.

2 Background

2.1 Particle attrition in fluidized beds

Particle attrition is a complex phenomenon. If a force is applied onto a particle it may break in two, a mechanism referred to as fragmentation. A lesser force may not be able to break the particle, but can give rise to a small number of coarse fragments instead (fines, dust etc), a mechanism referred to as abrasion. The daughter particles will have different characteristics compared to the mother particle, and may be subject to further attrition via different mechanisms. Particle attrition is affected by many variables, see Table 1.

Table 1: Some variables affecting attrition of particles.

Properties of particles	Properties of environment
Size	Time
Shape	Velocity
Surface	Pressure
Porosity	Shear
Hardness	Temperature
Cracks	

Fluidized bed reactors are an unforgiving environment, in which particles are subject to mechanisms that could cause high rate of mechanical attrition[8]. One source of attrition is the grid jets in the bottom of the bed, which typically involves nozzles with very high outlet gas velocities (above 100 m/s). Such devices are required to induce sufficient pressure drop to ensure good distribution of gas over the whole cross-section of the reactor. Fluidized beds usually also involves cyclones for separation of gas and solids, which give rise to attrition as the particles impact the cyclone walls at high speed. Other sources of attrition are collisions between particles when bubbling in the bed, or while splashing in the freeboard above the bed, see Table 2.

Table 2: Sources of mechanical attrition in circulating fluidized bed reactors.

Source	Description	Region of occurrence
Grid Jets	High velocity impact	Grid region
Cyclone	High velocity impact	Cyclone wall
Bubbling	Low velocity impact	Fluidized bed
Splashing	Low velocity impact	Freeboard

2.2 Particle attrition in chemical-looping processes

In chemical-looping processes, the oxygen carrier particles will not only be subject to mechanical forces, but also to chemical reactions, high temperature and sharp temperature gradients. This may induce attrition due to mechanisms such as stress or build-up of internal gas pressure. The situation is comparable to fluidized bed combustion with in-situ sulphur capture by limestone, in which the limestone particles are subject to mechanical and thermal forces as well as to chemical reactions[9]. Table 3 provides an overview of potential sources of attrition during chemical looping combustion in circulating fluidized beds, in addition to the mechanical sources listed in Table 2.

Table 3: Potential additional sources of attrition during chemical looping processes.

Factor	Description	Region of occurrence
Thermal shock	Induced stress, swelling, shrinking	Fluidized bed
Internal gas pressure	Sublimation, explosion	Fluidized bed
Chemical reaction	Change in crystal lattice, swelling, shrinking	Fluidized bed

At present, it is not possible to determine which of the factors suggested in Tables 2-3 are dominating in chemical-looping processes. The grid jets and cyclone seem very likely to be important. Hence reactor design could be expected to have considerable impact on mechanical attrition. The influence of the property change factors in Table 3 is currently not well understood and could be expected to differ considerably between different materials.

Currently, there are few studies examining particle attrition during chemical-looping combustion and related processes. A study with comprehensive methodology has been conducted by Brown et al.[10], studying iron ore and a CuO-based synthetic particle. Swelling and shrinking of oxygen carriers have been examined by Kimball et al.[11], examining one NiO and one CuO-based particle. Since these studies involve only two materials each, it is hard to draw general conclusions based solely on those. Others report attrition behaviour as complementary information to other results. For example, Gayan et al.[12] examined NiO-based particles by performing more than 120 cycles of oxidation and reduction in a bubbling fluidized bed, while reporting attrition as loss of fines per redox cycle. There are also studies concerning chemical-looping combustion in different kinds of reactor systems with continuous circulation of particles in which attrition behaviour has been examined, typically by examining changes in particle size distribution and determination of the amounts of fine material generated during operation, see for example Linderholm et al.[13], Berguerand et al.[14] and Adanez et al.[15]. The results presented below comes from a comprehensive study by the authors concerning measurement of attrition resistance of oxygen carrier particles with the jet cup method[16].

3 Experimental

3.1 Jet cup test rig

An experimental campaign to determine the resistance towards mechanical attrition for different oxygen carrier materials is presented. A customized test rig based on the jet cup method was used. While not a standardized method, the design allows for small samples and short test periods. A schematic description of the test rig can be found in Figure 1.

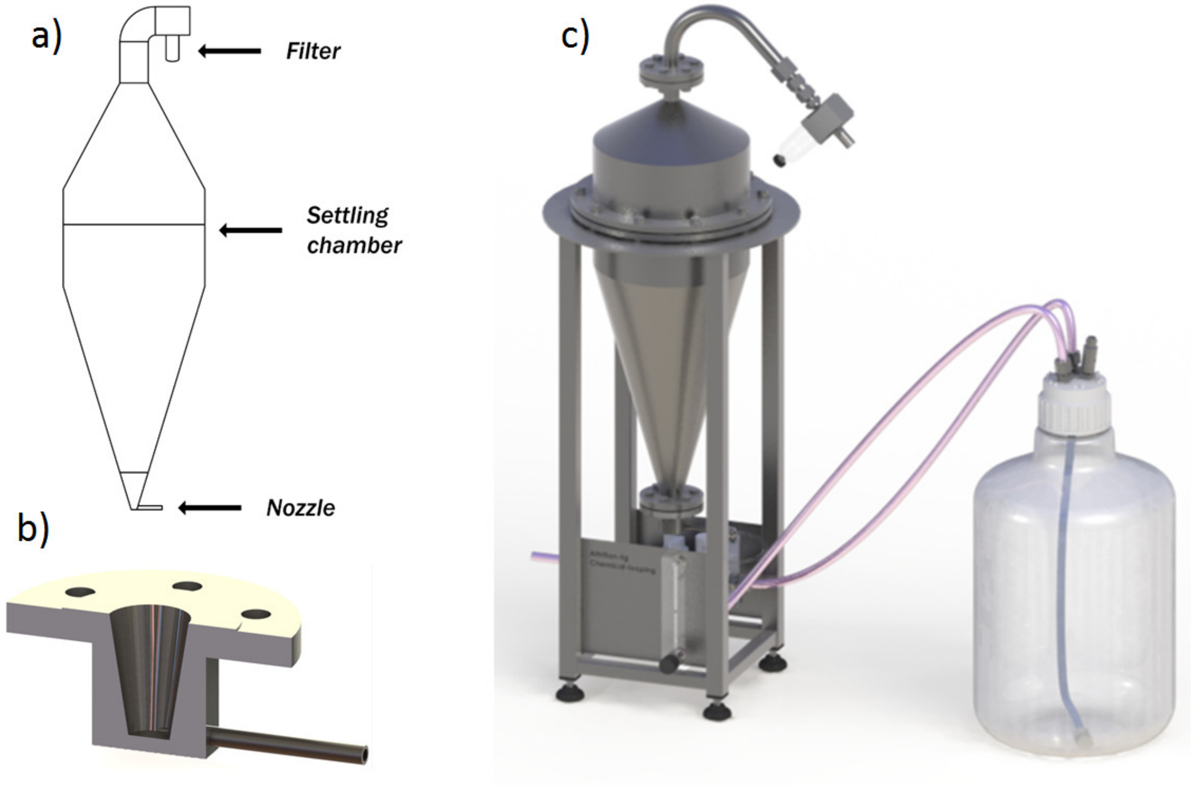

Figure 1: a) Schematic description of test rig, b) Geometry of the conical jet cup, c) Rendering of the whole rig

The apparatus consists of a 39 mm high conical jet cup with an inner diameter of 13 mm in the bottom and 25 mm in the top. A nozzle with an inner diameter of 1.5 mm is located at the bottom of the cup, and tangentially in relation to the cup wall. During operation, air is added with a velocity of 94 m/s through the nozzle, creating a vortex of particles swirling upwards. The design induces accelerated attrition by mechanisms similar to grid jets (due to high gas velocity at the exit of the nozzle) and cyclones (due to friction between wall and particles).

The jet cup is located at the bottom of a 634 mm high gravitational particle-gas separator, the settling chamber, basically a cone with a maximum diameter of 216 mm. Due to the increasing cross-section area, the gas velocity in the settling chamber is less than $^{1}/_{20000}$ compared to the inlet. The low gas velocity in the upper part of the gravitational separator allows elutriated particles to slow down and fall back into the cup, while generated fines are allowed to exit. A particle filter with a 0.01 µm filter element is located at the top of the device. An air humidifier is connected to the air inlet in form of a bottle in which air bubbles through a 240 mm high water column. The apparatus is operated at room temperature and close to atmospheric pressure.

3.2 Methodology

Prior to the experiments the filter is removed and weighed. The sample is prepared by sieving to 125-180 µm, or to 90-212 µm if insufficient amount is available in the more narrow size interval. The cup is then dismantled and a sample of 5 g of particles is added. The filter and the cup are then reattached to the apparatus. An air flow of 10 l/min is then applied to the nozzle, which corresponds to an air jet velocity of 94 m/s. The total test period is 1 h. The air flow is stopped once every 10 minutes in order to remove and weigh the filter. Although minor amounts of dust may stick to the cone walls in the upper part of the particle-gas separator, the increase of the filter's weight over the course of 60 min of testing should give a good indication of how much fines were produced during the period. Here the term fines refers to particles that are capable of passing the settling chamber and should consist mostly of particles with terminal velocity below the superficial gas velocity in the settling chamber, which is 0.0047 m/s. For most materials this equals particles smaller than ≈10 µm, but the exact cut-off diameter will be a function of density and geometry. After the test period, the apparatus is disassembled and the particles remaining in the cup collected and weighed.

Crushing strength was measured as the strength needed to fracture particles of the size 180-250 µm using a digital force gauge. The physical appearance and size distribution of the samples were determined by taking pictures with a light microscope. The images were then processed using a computer program calculating the size of each individual particle in each picture, approximated as the minimum Feret diameter. Many materials were also examined by sieving and by scanning electron microscopy (SEM), which could provide information about structural abnormities such as for example cracks in the surface of the particles.

3.3 Data evaluation

Different particles showed different behaviour in the jet cup. Most samples that had been subject to actual operation showed linear attrition as a function of time, see Figure 2a, while the majority of the fresh samples showed logarithmic attrition, such as in Figure 2b.

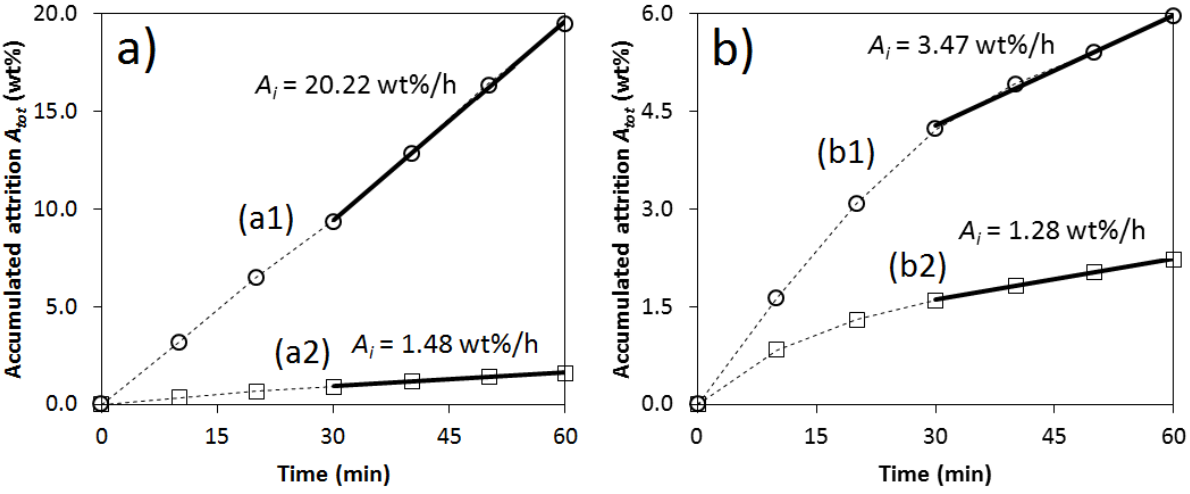

Figure 2: Example of attrition curve of used copper (a1) and iron (a2) based oxygen carriers showing linear attrition behavior, and of fresh copper (b1) and iron (b2) based oxygen carriers showing logarithmic attrition behavior. The "attrition index" A_i is defined as the slope of the curve for the last 30 min of the test.

Logarithmic attrition behaviour is not necessarily bad and was found to be common. Fresh particles often have small satellites attached to the main particle body, and spray-dried particles often contain hollow or doughnut-shaped particles. Once such weak particles, satellites and other irregularities have eroded, the remaining material typically was found to have high attrition resistance.

Among the samples were also a few made up of soft particles, most often which also had low density. During testing such particles often broke down into fragments that were too large to pass the gravitational particle-gas separator, but too small to fall back into the cup. Most of this material was stuck in the settling chamber even as the reactor was disassembled, and had to be removed by mild physical stimulation (i.e. hammer blows) or back blowing with air.

In order to be able to compare the performance of different samples, an "attrition index" A_i was defined as the slope of the curve for the last 30 min of the test, see equation 1, in which $m_{f,t}$ is the weight of the filter at a certain time during the test period and m_s is the initial weight of the particle sample.

$$A_i = 100 \times (60/30) \times (m_{f,t=60min} - m_{f,t=30min}) / m_s \qquad (wt\%/h) \qquad (1)$$

A_i provides the rate of attrition expressed as wt% fines caught in the filter over a certain time period. High values imply poor resistance to attrition.

It is important to point out that A_i is a product of a specific testing procedure, and should not be interpreted as the expected lifetime of oxygen carrier particles in a real-world chemical looping combustor. In fact, A_i could be expected to describe greatly accelerated mechanical attrition compared to what could be expected in an actual reactor due to the very harsh conditions in the jet cup. This can be exemplified by the expected life time of NiO-based particles produced by spray drying, for which the expected life time was estimated to 33000 h during extensive testing in a 10 kW_{th} chemical looping combustor[13], while A_i for the same particles measured with the jet cup indicates a lifetime of only 1250 h.

4 Results

4.1 Attrition resistance in jet cup and during continuous operation

All materials examined had previously been used as bed material in continuous operation. A complete summary of examined materials and their experimental history can be found elsewere[16]. In order to compare performance in the jet cup and performance in practical operation, each material was assigned to one of four groups (A-D) based on their performance. The groups are characterized by the following criteria:

A. Materials that have been successfully operated in 10 kW_{th} reactor with low attrition. The generation of fines was in all cases between 0.02-0.002 wt%/h, if defined as the amount of elutriated particles smaller than 45 µm. This reactor type involves riser gas velocities of about 3 m/s, cyclone inlet velocities around 8 m/s, and nozzle outlet velocities around 20 m/s, i.e. conditions comparable to large-scale applications.

B. Materials that have been successfully operated in 300 W_{th} reactor with low attrition. The generation of fines for these materials was below 0.05 wt%/h, if defined as the amount of particles smaller than 45 µm that are elutriated or present in the inventory after operation. This reactor type operates without cyclone and grid jets and all gas velocities are below 1 m/s.

C. Materials which have been successfully operated in 300 W_{th} reactor, but with more significant attrition compared to group B. For these materials the generation of fines was in the range 0.05-0.5 wt%/h with same definition of fines as in group B.

D. Materials that have experienced very significant attrition during operation. For these materials the formation of fines was greater than 0.5 wt%/h, using the definitions from above.

The attrition index and crushing strength of each sample divided into groups A-D can be found in Figure 3 below. Particles that experienced so severe fracturing that the test series could not be successfully concluded have been assigned an attrition index of 35. This was done in order for them to show up in the comparison even though it was not possible to measure a reliable attrition index for these materials.

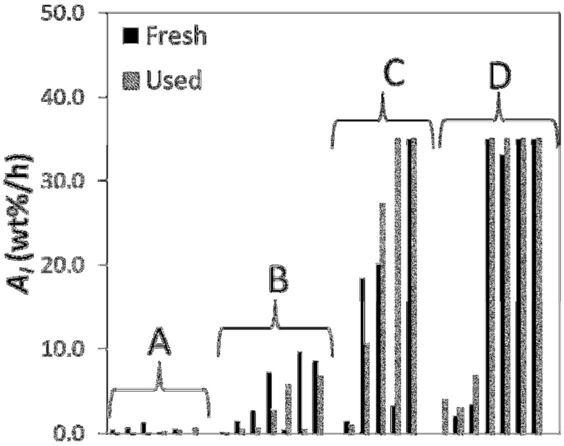

Figure 3a: Attrition index A_i of materials divided into groups A-D.

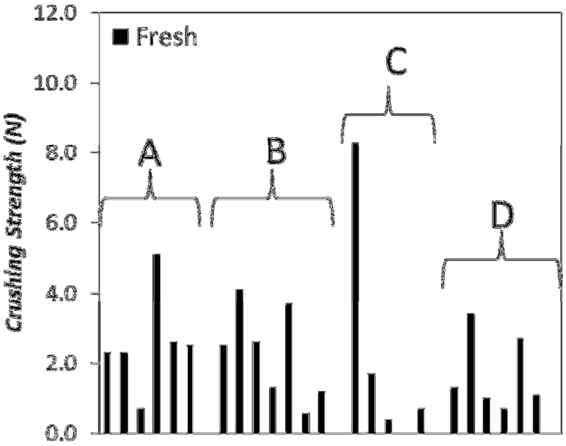

Figure 3b: Crushing strength of materials divided into groups A-D.

It is evident from Figure 3a that the attrition index gives a good indication of whether a material is suitable for continuous operation or not. Materials in group A and to some extent also those in group B clearly have lower attrition index compared to most of those in group C and D. This is true not only for used materials but also for fresh samples.

As for the commonly used crushing strength index, the correlation is not evident as can be seen in Figure 3b. However, it is reasonably clear that particles with a crushing strength above 2 N were much more likely to show good performance in actual operation.

It can be concluded that crushing strength, which is a quite simple method to use, may give a valuable first indication of whether a particle has sufficient attrition resistance or not. The jet cup method used in this work appears to provide a considerably better picture though. Finally, it should be stressed that sustained operation with fuel will always be necessary to draw safe conclusions, since jet cup tests take place at room temperature and without chemical reactions.

4.2 Performance of different oxygen carrier particles

The performance of different types of oxygen carriers are discussed in detail elsewere[16]. To summarize, the most important conclusions that can be drawn from the present study are that all composite materials with NiO or Fe_2O_3 as active phase and Al_2O_3-, $NiAl_2O_4$- or $MgAl_2O_4$-based support, as well as materials based on $CaMnO_{3-\delta}$ of perovskite structure, were found to have high attrition resistance, and it improved further following operation with fuel. In contrast, combined iron-manganese oxides, $(Fe_xMn_{1-x})_2O_3$, and materials containing smaller or larger amounts of either CuO or ZrO_2 experienced a reduction in attrition resistance during operation with fuel, and usually also had low attrition resistance to begin with. Crude materials such as manganese ore, ilmenite and iron oxide scales typically had decent attrition resistance.

4.3 Other observations

It was noted that there was always a considerable difference in attrition resistance between fresh and used particles of the same batch. This suggests that continuous oxidation and reduction of particles at high temperature affects the mechanical properties of oxygen carriers, and that performing measurements on fresh particles alone not necessarily is sufficient to judge whether a certain material is suitable for practical applications. The majority of the materials examined performed better after continuous operation with fuel.

Analysis of the particle size distribution of a few samples revealed that materials with low attrition resistance appear to have eroded mainly via fragmentation, while materials that performed better appear to have eroded via abrasion. This is illustrated in Figures 4-5.

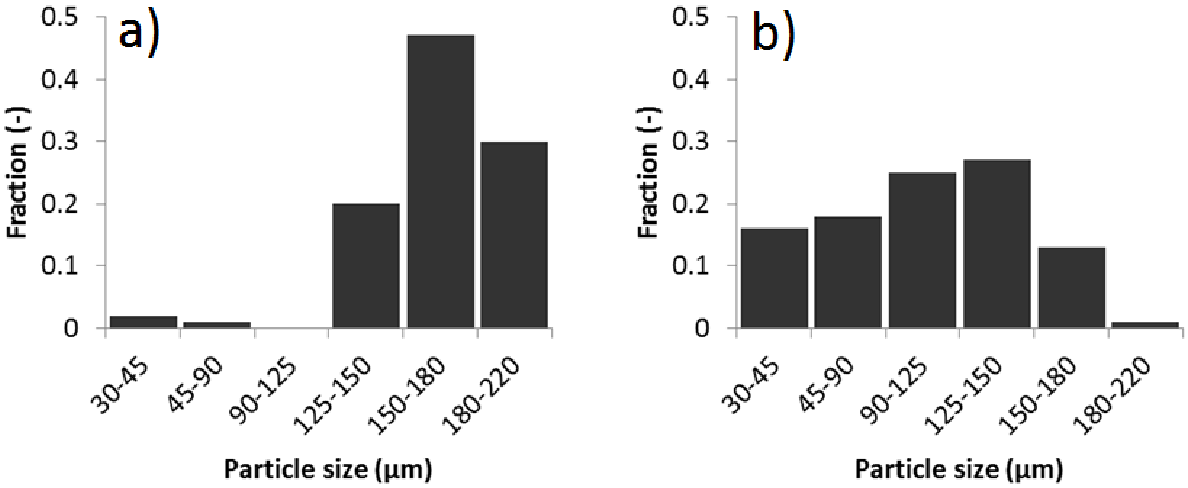

Figure 4: a) Particle size distribution for a poor performing material prior to jet cup test, b) particle size distribution for the same material after 1 h attrition test in jet cup.

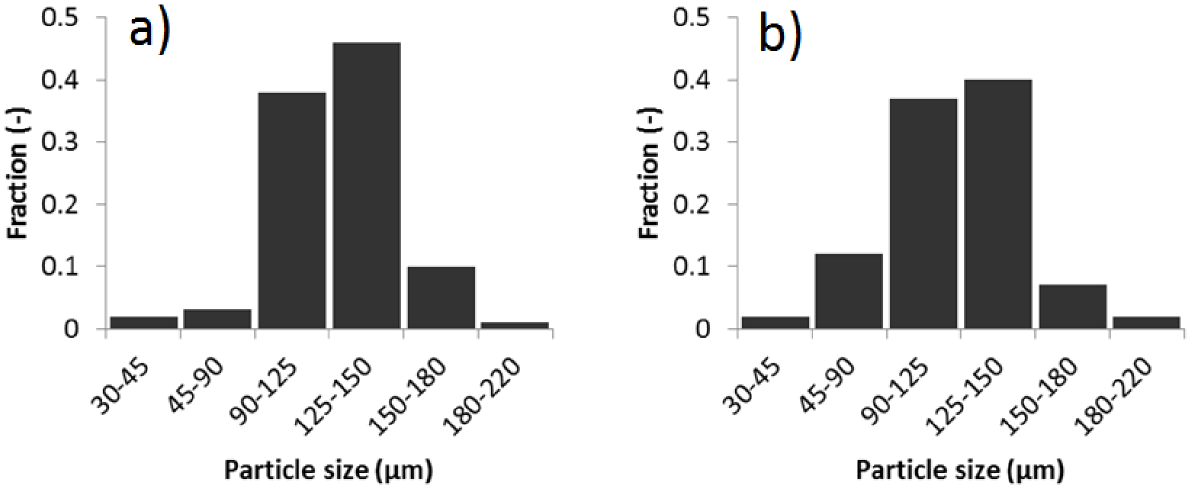

Figure 5: a) Particle size distribution for a good performing material prior to jet cup test, b) particle size distribution for the same material after 1 h attrition test in jet cup.

From Figure 4 it becomes evident that the poor performing material is fractured during attrition testing, since there is no other explanation for the presence of material in the size range 90-125 µm. In comparison, the particle size distribution of the good performing material did not change considerably, as can be seen in Figure 5. In this case the average particle size was only slightly reduced, which should be an effect of abrasion rather than fragmentation.

5 Discussion

It is clear that the jet cup rig used to measure attrition of oxygen carrier particles in this article has some limitations. Looking at the potential sources for particle attrition suggested in Table 2, it is evident that it takes at most two mechanisms into account (high velocity impact from grid jets and cyclone). Although these are expected to be the most important sources of mechanical attrition in chemical looping combustion, the methodology used does not take the effect of elevated temperature, internal gas pressure and chemical reactions into account. It should be noted though that it is common practice within the industry to measure attrition resistance of fluidized particles used at hot conditions in cold rigs, as is evident for example from the ASTM standard for FCC catalysts[17].

It is clear that the attrition index varies greatly between different materials. One may ask oneself if it would it be possible to establish an upper limit for acceptable attrition rate. That might be relevant for some applications. But as for chemical looping combustion it should be kept in mind that the research field is currently wide open. At this point in time it is possible to imagine the use of both expensive materials with long life time and cheap materials with short life time. The choice may turn out to be different for different applications. Coal combustion may require cheap and easily replaceable oxygen carriers that are resistant to sulphur and ash, while natural gas can be cleaned from impurities so that high performing but more expensive oxygen carriers could be favourable. Therefore attrition resistance should be seen as one factor of many when considering the performance of oxygen carrier materials.

While this study indicates a quite robust correlation between crushing strength, jet cup attrition tests and results from actual operation there are also some important deviations. This may be seen as a weakness of the method, but it should be pointed out that the lack of complete correlation also provides important information about areas where the knowledge needs to be improved. These differences may in fact contribute to improved understanding of attrition mechanisms. If, for example, a certain type of material performs well in jet cup attrition testing but very poorly in actual operation this could be an indication that chemical reactions or high temperature is of prime importance for such materials.

6 Conclusions

Basic theory concerning attrition of oxygen carrier material has been discussed, and the results from attrition tests and crushing strength tests for a number of different materials that previously have been subject to continuous chemical looping experiments are briefly presented. The following conclusions can be drawn:

- Materials that have performed well during continuous operation at elevated temperature and continuous redox reactions typically also performed well during jet cup tests. Therefore it is clear that jet cup tests at room temperature do provide a meaningful indication concerning the feasibility of different oxygen carrier materials.
- No strong correlation between the commonly used crushing strength index and attrition resistance measured with the jet cup described in this paper has been found. However, it was clear that particles with a crushing strength above 2 N were more likely to perform well, compared to softer particles.
- Composite materials with NiO or Fe_2O_3 as active phase and Al_2O_3-, $NiAl_2O_4$- or $MgAl_2O_4$-based support, and materials based on $CaMnO_{3-\delta}$ of perovskite structure, were found to have high attrition resistance. In contrast, combined $(Fe_xMn_{1-x})_2O_3$ oxides and materials containing smaller or larger amounts of either CuO or ZrO_2 performed poorly.

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8 References

[1] Boot-Handford M.E., Abanades J.C., Anthony E.J., et al. (2014), Carbon capture and storage update, *Energy & Environmental Science* **7**, pp. 130-189

[2] Rydén M., Lyngfelt A., Mattisson T. (2006), Synthesis gas generation by chemical-looping reforming in a continuously operating laboratory reactor, *Fuel* **85**, pp. 1631-1641

[3] Lind F., Seemann M., Thunman H. (2011), Continuous catalytic tar reforming of biomass derived gas with simultaneous catalyst regeneration, *Industrial & Engineering Chemistry Research* **50**, pp. 11553-11562

[4] Rydén M., Lyngfelt A. (2006), Using steam reforming to produce hydrogen with carbon dioxide capture by chemical-looping combustion, *International Journal of Hydrogen Energy* **31**, pp. 1271-1283

[5] Rydén M., Arjmand M. (2012), Continuous hydrogen production via the steam-iron reaction by chemical looping in a circulating fluidized-bed reactor, *International Journal of Hydrogen Energy* **37**, pp. 4843-4854

[6] Thunman H., Lind F., Breitholtz F., et al. (2013), Using an oxygen-carrier as bed material for combustion of biomass in a 12-MWth circulating fluidized-bed boiler, *Fuel* **113**, pp. 300-309

[7] Adanez J., Abad A., Garcia-Labiano F., et al. (2012), Progress in Chemical-Looping Combustion and Reforming technologies, *Progress in Energy and Combustion Science* **38**, pp. 215-282

[8] Werther J., Reppenhagen J. (1999), Catalyst attrition in fluidized-bed systems, *AIChE Journal* **45**, pp. 2001-2010

[9] Scala F., Salatino P., Boerefijn R., et al. (2000), Attrition of sorbents during fluidized bed calcination and sulphation, *Powder Technology* **107**, pp. 153-167

[10] Brown T.A., Scala F., Scott S.A., et al. (2012), The attrition behaviour of oxygen-carriers under inert and reacting conditions, *Chemical Engineering Science*, **71**, pp. 449-467

[11] Kimball E., Lambert A., Fossdal A., et al. (2013), Reactor choices for chemical looping combustion (CLC) dependencies on materials characteristics, *Energy Procedia*, **37,** pp. 567 - 574

[12] Gayán P., de Diego L.F., García-Labiano, F., et al. (2008), Effect of support on reactivity and selectivity of Ni-based oxygen carriers for chemical-looping combustion, *Fuel* **87**, pp. 2641-2650.

[13] Linderholm C., Mattisson T., Lyngfelt A. (2009), Long-term integrity testing of spray-dried particles in a 10-kW chemical-looping combustor using natural gas as fuel, *Fuel* **88**, pp. 2083-2096.

[14] Berguerand N., Lyngfelt A. (2008), The use of petroleum coke as fuel in a 10 kW_{th} chemical-looping combustor, *International Journal of Greenhouse Gas Control* **2**, pp. 169-179.

[15] Adánez J., Gayán P., Celaya J., et al. (2006), Chemical looping combustion in a 10 kW_{th} prototype using a CuO/Al_2O_3 oxygen carrier: Effect of operating conditions on methane combustion, *Industrial Engineering Chemistry Research* **45**, pp. 6075-6080.

[16] Rydén M., Moldenhauer P., Lindqvist S., et al. (2014), Measuring attrition resistance of oxygen-carrier particles for chemical-looping combustion with a customized jet cup, *Powder Technology* **256**, pp.75-86.

[17] ASTM D5757-95: Standard Test Method for Determination of Attrition and Abrasion of Powdered Catalysts by Air Jets; ASTM: Philadelphia, United States, 1995.