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# Development and testing of ilmenite granules for packed bed chemical-looping combustion

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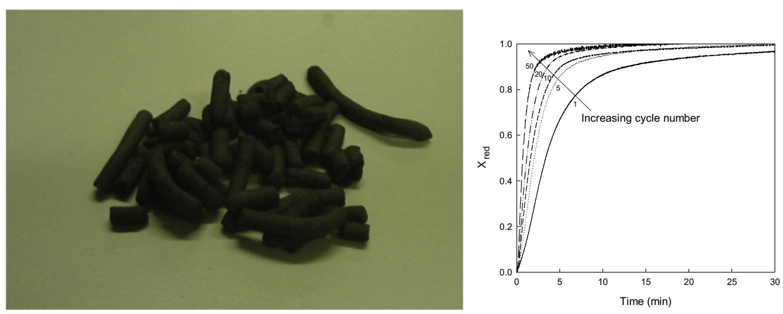
## HIGHLIGHTS

- Different particles have been developed and tested for application in a packed-bed CLC reactor.
- Ilmenite was chosen as base material due to its good reactivity with syngas, the natural availability and related low cost.
- A suitable oxygen-carrier was developed using ilmenite as base material and $Mn_2O_3$ as additive.
- No significant differences in the reduction and oxidation reactivity of the different granules were observed.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Chemical-looping combustion (CLC) is a promising technology that integrates power production and $CO_2$ capture with a low energy penalty. CLC has been successfully demonstrated using interconnected fluidized bed reactor systems. However, high pressure operation allows the use of inherently more efficient power cycles than low pressure fluidized bed solutions. With the aim to work at elevated pressures, dynamically operated packed-bed reactors have been proposed for CLC.

In a packed-bed CLC reactor bigger oxygen carrier particles are used to avoid very large pressure drops and the required mechanical properties of these particles are quite different from the properties needed in a fluidized bed CLC system. In this work different particles have been developed and tested for application in a packed-bed CLC reactor. Ilmenite was chosen as base material because of its good reactivity with syngas, the natural availability and related low cost. Different ilmenite pellets with different composition and shape were developed and their mechanical properties before and after thermal and chemical cycling were analysed and compared. The reactivity of the particles and the influence of different reaction conditions were also studied in a thermogravimetric analyser. It was found that the thermal and chemical stresses produced an important deterioration of the mechanical properties of the pellets. The type of additive used during the production process showed an important effect on the mechanical properties of the granules. Only the granules produced with $Mn_2O_3$ as additive demonstrated acceptable suitable mechanical properties after thermal and chemical cycling.

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**Nomenclature**

| | | | |
|---|---|---|---|
| $C_p$ | heat capacity (kJ kg$^{-1}$ K$^{-1}$) | 0 | initial condition |
| $\Delta H_R$ | heat of reaction (kJ mol$^{-1}$) | *act* | active metal |
| $M_i$ | molecular weight specie *i* (kg mol$^{-1}$) | *g* | gas phase |
| $\Delta T$ | temperature change (°C) | *i* | component in the gas phase |
| $\zeta$ | stoichiometric factor | *ox* | oxidized state |
| $\omega$ | weight fraction | *red* | reduced state |
| | | *s* | solid phase |

*Sub- and superscripts*

## 1. Introduction

The increase in the $CO_2$ concentration in the atmosphere mainly due to the combustion of fossil fuels has contributed to the global warming problem [1]. There are different ways to decrease anthropogenic $CO_2$ emissions: reducing energy consumption by increasing the efficiency of energy conversion and/or utilization; switching to less carbon intensive fuels; increasing the use of renewable or nuclear energy; and the capture and storage of $CO_2$ in geological formations (CCS). According to the IPCC and IEA [2,3], CCS could account for 19% of the total $CO_2$ emission reductions needed this century to stabilize climate change at a reasonable cost. Therefore, the development to market maturity of CCS technologies is essential for the long-term development of clean energy both to ensure a continued role of fossil fuels, in particular coal, as well as to reduce global emissions.

There are different technologies available or currently under development which can accomplish the capture of $CO_2$ from combustion sources, i.e. pre-combustion, post-combustion or oxyfuel technologies. Nowadays, most of these processes require high amounts of energy resulting in an increase in the cost of energy production and efficiency losses.

Chemical looping combustion (CLC) has been proposed [4,5] as a possible solution for the disadvantages of other capture technologies, because it combines power production and $CO_2$ capture in a single stage and produces a pure $CO_2$ stream ready for compression and sequestration without any separation step or need for additional energy. A solid oxygen carrier in the form of metallic oxide particles transports the oxygen from the air to the fuel, thus avoiding the dilution of exhaust gases from the power plant with $N_2$.

The process takes place in cycles of two steps. In the first step (reduction step) the fuel is oxidized to $CO_2$ and $H_2O$ by a metal oxide (MeO) that is reduced to a metal (Me) or a reduced form of MeO. In the second step (oxidation step) the metal or reduced oxide is oxidized with air, and the material is regenerated and ready to start a new cycle. The flue gas leaving the oxidation step contains $N_2$ and unreacted $O_2$ at high temperatures which is sent to a gas turbine for electricity production. The exit gas during the reduction step contains only $CO_2$ and $H_2O$. After water condensation, almost pure $CO_2$ can be obtained with little energy lost for $CO_2$ purification. The total amount of heat evolved from the reactions in the two steps is the same as for conventional combustion, where the oxygen is in direct contact with the fuel.

The method used to transfer the oxygen between the air and fuel, depends on the reactor configuration. The configuration composed of two interconnected fluidized-bed reactors working at atmospheric pressure has been widely studied and demonstrated at different scales [6–8]. However, the efficiency of power cycles increases at elevated temperatures (>1200 °C) and pressures (15–20 bar) and in this respect, operation of pressurized CLC plants at very high temperatures based on interconnected fluidized bed technology could pose technical difficulties especially due to the gas/solid (fines) separation. With the aim to work at elevated pressures, dynamically operated packed-bed reactors have been proposed for CLC [9–11]. At least two reactors in parallel working in alternating cycles must be used to assure a continuous high temperature gas stream supply to the downstream gas turbine (see Fig. 1). The main advantages of the packed-bed reactor technology are that the separation of gas and particles is intrinsically avoided, the easiness in design and scale up and the possibility to work under elevated pressure. A disadvantage of this concept includes the necessity to use high temperature gas switching valves.

In both CLC configurations, the key issue for the system performance is the selection of an oxygen carrier with suitable properties, such as sufficient oxygen transport capacity, high reactivity under alternating reducing and oxidizing conditions to reduce solids inventory, very good chemical and mechanical stability, resistance to attrition to minimize losses of elutriated solids, high selectivity to $CO_2$ and $H_2O$, and low tendency for carbon deposition. Other important requirements are high availability and low cost of the metal, as well as low environmental impact.

Recently, the development of oxygen carriers has focused on these last two properties. Different industrial waste products, ores and minerals have been tested as possible oxygen carriers [12–14].

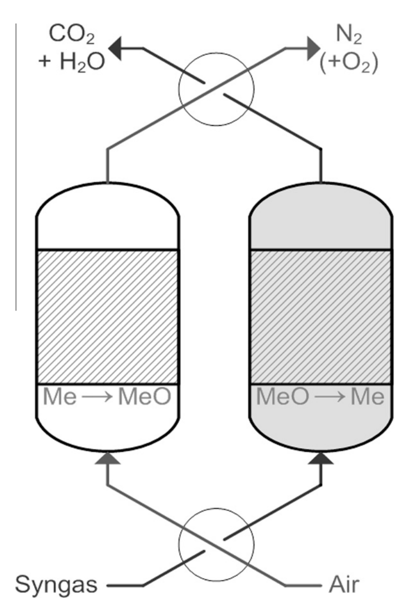


**Fig. 1.** Schematic diagram of CLC in packed-bed reactors.

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