



On the use of a highly reactive iron ore in Chemical Looping Combustion of different coals



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HIGHLIGHTS

- A highly reactive OC (Tierga iron ore) was tested in a continuous 500 W_{th} unit.
- Lower oxygen demand than ilmenite and Fe-ESF was obtained regardless the temperature.
- Using a carbon separation unit, carbon capture efficiencies >95% are expected.
- CaO in the OC allowed SO₂ retention but too high makeup flows would be required.
- Tierga iron ore maintained chemical and physical properties after 50 h operation.

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ABSTRACT

Coal combustion using the Chemical Looping technology can be carried out under different configurations. This paper focuses on the *in situ* gasification Chemical Looping Combustion (iG-CLC). In this technology, it is especially important the selection of the oxygen carrier as there may be losses in the drainage of coal ashes. Finding low-cost oxygen carriers has become a relevant research focus. Several Fe-based materials have been tested including minerals and industrial residues. In this work, a highly reactive iron ore that had already shown promising characteristics for coal combustion was used in a continuous 500 W_{th} CLC unit. Its performance in the combustion of anthracite, bituminous coal and lignite was evaluated and compared with the results for other Fe-based materials, such as ilmenite or a bauxite waste. The combustion efficiency obtained with the Tierga iron ore was the highest reported to date which makes this carrier a promising candidate for further scale-up. Moreover, the high CaO content of this material led to analyze its relevance for sulphur removal during the first hours of operation. High sulphur retention capacity was observed but this capacity decreased with time as the calcium oxide was both saturated and lost as fines during operation.

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1. Introduction

According to IEA statistics, carbon dioxide emissions from fossil fuel combustion for heat and power production accounted for 41% of world CO₂ emitted in 2008 [1]. The principal fuel used in this sector is coal, the most carbon-intensive of fossil fuels and its use is expected to become even more widespread in certain world regions. In order to mitigate the impact of CO₂ emissions on climate change, CO₂ Capture and Storage (CCS) emerged as an option to reduce CO₂ emissions from the power sector by separating CO₂ from the rest of combustion gases and storing it in a safe place. Thus CO₂ emission to the atmosphere is avoided. Among the different CO₂ capture technologies already investigated, this paper focuses on

Chemical Looping Combustion (CLC). In this technology, the combustion of the fossil fuel takes place avoiding the direct contact between fuel and air. The presence of an oxygen carrier, normally a metal oxide, allows for the transfer of oxygen from air to fuel by means of a redox reaction. The oxygen carrier circulates between two reactors, called fuel and air reactors. The oxygen carrier is reduced in the fuel reactor and the oxygen transferred in this process oxidizes the fuel to CO₂ and H₂O. The reduced oxygen carrier is then conveyed to the air reactor, where it is oxidized back to the initial state. Then, the oxygen carrier is sent again to the fuel reactor to start a new redox cycle allowing the continuous combustion of the fuel.

Different configurations have been described for CLC of coal, but most of them consider interconnected fluidized beds for fuel and air reactors. This paper is based on the *in situ* Gasification Chemical Looping Combustion (iG-CLC) [2]. According to the iG-CLC scheme,

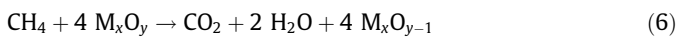
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Nomenclature

C_{fixed}	percentage of fixed carbon (%)	p_{react}	partial pressure of the gaseous reactants (bar)
C_{coal}	carbon percentage in coal (%)	$(-r_C)$	rate of char conversion (s^{-1})
$F_{CO_2,AR}$	carbon dioxide molar flow in the air reactor (mol/s)	$(-r_O)$	rate of oxygen transferred from the oxygen carrier to the fuel (mol/s)
$F_{i,FR}$	i species molar flow in the fuel reactor inlet/outlet stream (mol/s)	R_{OC}	oxygen transport capacity
$F_{C,vol}$	carbon flow from the volatile matter (mol/s)	t_{res}	residence time in the fuel reactor (s)
F_{OC}	Solid circulation rate (kg/s)	X_{char}	char conversion
K_i	constant considering the inhibitory effect in Eq. (16)	X_r	Oxygen carrier conversion during reduction
k_{react}	kinetic constant in Eq. (16)	<i>Greek symbols</i>	
M_O	molar mass of oxygen (0.016 kg/mol)	ϕ	oxygen carrier to fuel ratio
M_{O_2}	molar mass of molecular oxygen (0.032 kg/mol)	Ω_{SF}	coal oxygen demand (kg oxygen/kg coal)
\dot{m}_{OC}	solid circulation rate (kg/s)	Ω_T	total oxygen demand
\dot{m}_{SF}	coal feeding rate (kg/s)	η_{CC}	carbon capture efficiency
$m_{OC,FR}$	mass of oxygen carrier in the fuel reactor (kg)	η_{CC}^*	carbon capture efficiency considering elutriation
$m_{s,FR}$	mass inventory in the fuel reactor (kg oxygen carrier/ MW_{th})	η_{CS}	carbon separation efficiency in the carbon separation unit
p_{prod}	partial pressure of the gasification products (CO and H ₂) (bar)		

coal is introduced directly to the fuel reactor where it is gasified. Steam or CO₂ or even mixtures of both are commonly used as gasifying agents. The following equations summarize the processes taking place:



The reduced oxygen carrier is then transferred to the air reactor where it is re-oxidized again:



The performance of the CLC technology can be evaluated by the carbon capture and combustion efficiency of the process. High values of carbon capture efficiency indicate that most of the carbon in the coal fed can be found in the gaseous stream at outlet of the fuel reactor and therefore captured. In order to maintain high carbon capture values, the transfer of carbon from fuel to air reactor should be minimized as the carbon dioxide emitted in the air reactor is not captured. Therefore, the residence time of char in the fuel reactor should be long enough for the char to be gasified [3–5]. To improve char conversion maintaining a reasonable reactor size, the use of a carbon separation unit was proposed [6], where unconverted char is separated from oxygen carrier particles and sent back to the fuel reactor.

High combustion efficiency values are related to the absence of unburned compounds at the outlet of the fuel reactor. The combustion efficiency in *i*G-CLC is evaluated using the oxygen demand, which indicates the fraction of oxygen demanded by unconverted compounds exiting from the fuel reactor (CH₄, H₂ or CO) with respect to the oxygen required to burn the solid fuel. The oxygen demand would be directly related to the O₂ requirements in a hypothetical oxygen polishing step downstream the fuel reactor [7]. The presence of unconverted compounds in *i*G-CLC has been reported before in units ranging 0.5–100 kW_{th} with different oxy-

gen carriers and solid fuels [8–17] and represents both energy and CO₂ quality losses [18]. Recent investigations try to decrease the oxygen demand of the process, which includes optimization of operational conditions, design modifications or use of highly reactive materials [18–21]. Recently, a summary of experimental data obtained in different experimental units under different combustion conditions was presented by Gayán et al. [19]. The analysis of the data allowed them to evaluate the impact of the type of solid fuel, the oxygen carrier material and the solids inventory on the oxygen demand. The majority of the results that were considered used ilmenite as oxygen carrier. Ilmenite is a mineral found in metamorphic and igneous rocks. The principal constituent of ilmenite is FeTiO₃. The oxygen demand values reported for ilmenite under different experimental conditions varied between 5% and 15%. Experimental results obtained in different CLC units with different oxygen carriers and different coals suggest that the use of oxygen carriers with higher reactivity than ilmenite (based on Ni or Fe) allowed reaching lower values of oxygen demand for the same solids inventory in the fuel reactor. Also, it was found that solid fuels with an important volatile content usually showed higher oxygen demand values. Theoretical calculations confirmed that the oxygen carrier reactivity notably influenced the oxygen demand [19,20].

On the other hand, being cost-competitive and easy to dispose are two features that become especially significant for an oxygen carrier in an *i*G-CLC system burning coal. Depending on the ash content of the coal, it could be necessary to periodically drain the coal ashes from the CLC system. Otherwise, they would accumulate and generate operational problems. Therefore some oxygen carrier particles can be lost together with the ashes during this process and that is the reason to look for cheap and environmentally friendly materials. According to this, Fe-based materials represent a more interesting option for *i*G-CLC of coal. Although they are not as reactive as metal oxides like NiO, they present the advantage of their low cost and non-toxic nature [2]. In many recent studies the oxygen carriers tested in *i*G-CLC units with coal were industrial residues and minerals [17,22,23].

In the research group at Instituto Carboquímica ICB-CSIC different Fe-based materials have been tested as oxygen carriers for *i*G-CLC. Those tests included chemical and physical characterization of ilmenite, a bauxite waste generated in the alumina production through the Bayer process (Fe-ESF), an iron ore from a hematite mine in Tierga (Zaragoza, Spain) and a copper ferrite ore [24,25].

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