



Evaluation of the use of different coals in Chemical Looping Combustion using a bauxite waste as oxygen carrier

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HIGHLIGHTS

- ▶ A bauxite waste (BW) was identified as a promising oxygen carrier for *i*G-CLC of coal.
- ▶ BW showed high combustion efficiencies of gasification products using different coals.
- ▶ The use of recycled CO₂ as gasifying agent should be limited except for lignite.
- ▶ In TGA, BW-char reaction extent was larger for chars from low rank coals.
- ▶ In fluidized bed, no significant evidence of BW-char reaction was observed.

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ABSTRACT

The interest in the use of solid fuels such as coal in Chemical Looping Combustion (CLC) is growing because of the benefits of the direct use of coal in this technology on the reduction of the costs linked to carbon dioxide capture. In CLC, the oxygen needed for the combustion is supplied by a solid oxygen carrier therefore avoiding the direct contact between fuel and air. Focusing on the use of solid fuels in the *in situ* Gasification Chemical Looping Combustion (*i*G-CLC), the oxygen carrier is mixed with the coal in the fuel reactor, where gasification of the coal and reaction of the gasification products with the oxygen carrier particles take place. In this process, the possible loss of oxygen carrier together with the ashes makes it interesting to find inexpensive materials to be used as carriers, such as natural minerals or industrial residues. In this work, an Fe-based bauxite waste is used as oxygen carrier in the combustion of different types of coal. Experiments were performed in a TGA and a batch fluidized bed (FB) using both coal and char from the corresponding coal. The influence of temperature as well as the gasifying agent composition on the performance of coal conversion in an *i*G-CLC process was evaluated. Tests in a thermogravimetric analyzer (TGA) revealed direct char combustion by oxygen in the bauxite waste material, but no evidences of such direct combustion were found in the experiments in the batch fluidized bed. In this case, gasification of char by H₂O or CO₂ was found as a necessary step in char conversion. Using steam as gasifying agent, higher char gasification rates were observed than using CO₂ and in all conditions lignite presented the highest char gasification rate. At 980 °C, the lignite char gasification rate in CO₂ doubled the value obtained with anthracite using steam, indicating that recirculated CO₂ can be fed to the fuel reactor of a CLC system if lignite is used as fuel. For the rest of the fuels, it was possible to use a mixture of H₂O and CO₂ as gasifying agent without decreasing the feasibility of the *i*G-CLC process. The bauxite waste was able to burn the gaseous products generated during the gasification of the different types of coal with high combustion efficiencies.

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1. Introduction

The increase in the atmospheric carbon dioxide concentration leads to an increased global temperature [1]. According to recent estimations, fossil fuels will continue contributing with an important share to fulfill the energy generation requirements, thus

increasing the anthropogenic CO₂ emitted to the atmosphere [2]. The use of coal is expected to remain in an important position in the energy mix in the foreseeable future as it is more abundant and less expensive than natural gas [3].

One of the possibilities to reduce CO₂ from fossil fuel combustion that is receiving increasing interest is Carbon Capture and Storage (CCS) which focuses on storing CO₂ at site, such as depleted oil/gas fields or deep saline aquifers, thus preventing that CO₂ from reaching the atmosphere. Among the CO₂ capture alternatives

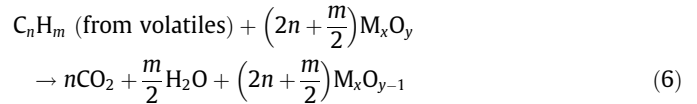
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Nomenclature

C_c	carbon content of the char (ash free)	O/C	oxygen to carbon molar ratio
ε	decrease percentage of the average char conversion rates for steam and CO ₂ mixtures	$\Omega_g(t)$	oxygen demand of the gas products at time t (%)
$F_{CO_2,in}$	CO ₂ flow introduced in the fuel reactor (mol/s)	$\Omega_{g,av}(t)$	averaged oxygen demand of the gas products during the reducing period (%)
$F_{H_2O,in}$	H ₂ O flow introduced in the fuel reactor (mol/s)	R_{OC}	oxygen transport capacity
$F_{H_2O,out}$	H ₂ O flow in the product gas (mol/s)	$r_c(t)$	rate of fuel conversion (mol/s)
F_i	molar gas flow of each component i (CO ₂ , CO, H ₂ or CH ₄) in the product gas (mol/s)	$r_{c,inst}$	instantaneous rate of fuel conversion (s ⁻¹)
F_{N_2}	downstream introduced N ₂ flow (mol/s)	$r_{O(t)}$	rate of oxygen transferred from the bauxite waste to the fuel gas (mol/s)
$F_{O,in}$	oxygen flow introduced (mol O/s)	t	time (s)
$F_{O,out}$	oxygen flow at the reactor outlet (mol O/s)	t_{95}	time to convert 95% char (s)
F_{out}	dry outlet flow after nitrogen dilution (mol/s)	t_x	residence time to reach a conversion (s)
m	mass weight variation in the TGA experiments (Kg)	X_{char}	char conversion
M_O	oxygen molar weight (kg/mol)	X_{coal}	coal conversion
M_xO_y	oxidized form of the oxygen carrier	X_{red}	bauxite waste conversion for the reduction reaction being the molar fraction of the component i (CO ₂ , CO, H ₂ or CH ₄) in the product gas
M_xO_{y-1}	reduced form of the oxygen carrier	y_i	
m_o	mass of the oxidized form of the oxygen carrier (kg)	η_c	combustion efficiency of the gasification products
m_r	mass of the reduced form of the oxygen carrier (kg)		
m_{OC}	mass of the oxygen carrier (kg)		
$N_{O,bw}$	moles of oxygen in the bauxite waste active for CLC process (mol)		
$N_{C,char}$	moles of carbon from char fed into the reactor (mol)		
$N_{C,coal}$	moles of carbon from coal fed into the reactor (mol)		
$O_{2,demand,coal}$	oxygen needed for complete combustion of the coal fed		
		Acronyms	
		OC	oxygen carrier
		TGA	thermogravimetric analyzer
		A	anthracite
		HVB	bituminous with high volatile content
		MVB	bituminous with low volatile content
		L	lignite

currently in development, the Chemical Looping Combustion (CLC) technology has emerged as a promising option to capture CO₂ at low cost [4]. The main idea behind CLC is to avoid CO₂ dilution in the exhaust gases by providing the oxygen needed for combustion by means of an oxygen carrier, normally a metal oxide, so that a concentrated CO₂ stream is obtained. The preferred configuration for CLC is two interconnected fluidized bed reactors, fuel reactor and air reactor with the oxygen carrier circulating between them [5,6]. Different approaches for solid fuel combustion have been developed until now [6]. This work focuses on the *in situ* Gasification Chemical Looping Combustion (iG-CLC) technology for solid fuels. This technology was recently demonstrated during continuous operation in CLC units up to 10 kW_{th} [7–9]. The scheme of the process is shown in Fig. 1. In the iG-CLC process, the coal is introduced directly to the fuel reactor and mixed with the oxygen carrier, where it is devolatilized to produce volatile matter and solid char (reaction (1)). Char, which is mainly composed by carbon and ash, is gasified using steam or CO₂ continuously fed to the fuel reactor, following reactions (2) and (3). Several previous studies have pointed to char gasification as the rate limiting step [10–13]. After gasification, the gaseous products react with the oxygen carrier present in the fuel reactor to produce water and CO₂. These processes are summarized in reactions (4)–(6). Once water has been condensed, a highly-pure CO₂ stream is obtained ready for capture. The reduced oxygen carrier is then transferred to the air reactor where it is regenerated (reaction (7)) and sent back to the fuel reactor to start a new redox cycle.



In experiments performed in a continuous iG-CLC unit, unburnt gases (CH₄, CO and H₂) in the fuel reactor exit were measured using a bituminous coal as fuel and ilmenite as oxygen carrier [8]. Nevertheless, this study showed that these gases were volatile matter that had not been fully oxidized by ilmenite. Different alternatives have been proposed to deal with the unburnt gases, e.g. an oxygen polishing step downstream to complete gas combustion to CO₂ and H₂O, separation and recirculation of unburnt compounds or fuel reactor in series where exhaust gases were fed. It is expected that the use of little fraction of the total solids in the system in this second reactor had a high impact on the combustion efficiency increase [14].

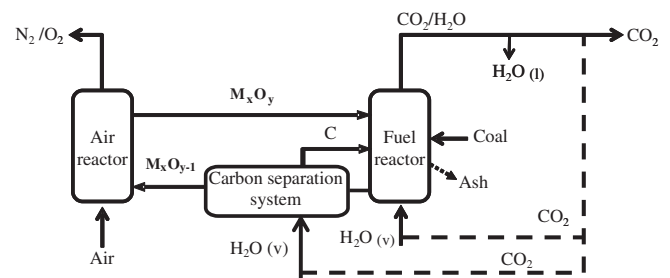


Fig. 1. *In situ* Gasification Chemical Looping Combustion process. (---) Optional streams to replace H₂O (v) by CO₂ in the fluidizing gas of fuel reactor and/or the carbon separation system.

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