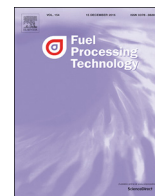




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Research article

Assessment of the improvement of chemical looping combustion of coal by using a manganese ore as oxygen carrier

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ABSTRACT

Finding suitable low-cost materials to be used as oxygen carrier in Chemical Looping Combustion (CLC) of coal is a key issue to achieve the CO₂ capture at low economic cost. Recently, a Mn-ore from Gabon has been identified as an alternative to the state of the art of oxygen carriers based on minerals or wastes with high iron content. This Mn-ore showed a high reactivity and a long particle lifetime during batch characterization to be considered as a suitable oxygen carrier. To evaluate the potential of this material in CLC, this work analyses the behaviour of the Mn-ore during continuous combustion of a bituminous coal in a 0.5 kW_{th} CLC unit. The CLC process was evaluated and the effect of the main operating variables - such as fluidizing medium, oxygen carrier circulation rate, temperature, and solids inventory in the fuel reactor - on the combustion efficiency and CO₂ capture was investigated. A direct relation between the char conversion rate and the CO₂ capture is given, being mainly affected by the mean residence time of solids and temperature in the fuel reactor. The use of a carbon separation system with separation efficiency above 90% would be required to achieve CO₂ capture rates higher than 95%. Total oxygen demand values as low as 4.5% were found when optimal operating conditions were selected, mainly being related to oxygen carrier to fuel ration higher than $\phi > 3$. At these conditions, the Mn-ore material showed similar combustion efficiencies than other Fe-based low-cost materials previously tested, but with higher CO₂ capture rates.

1. Introduction

New technologies are being investigated to reduce the CO₂ emissions to the atmosphere by fossil fuel combustion. Among them, Chemical Looping Combustion (CLC) has been highlighted as a promising one with low economic and energetic costs [1]. CLC is based in two reactor so-called fuel and air reactors with an oxygen carrier continuously circulating between them. In the fuel reactor, the fuel combustion happens using oxygen available in the oxygen carrier, usually being a metal oxide. In the air reactor, the oxygen carrier is regenerated by oxidation with air. In this way, air is not put in contact with the fuel, allowing the inherent CO₂ capture in the process, while the global chemical reaction and combustion enthalpy is the same than in normal combustion with air.

The development of the CLC with solid fuels has been mainly focused on the in-situ Gasification (iG-CLC) concept [2–4], where steam and/or recycled CO₂ are supplied to the fuel reactor as gasifying agents. A scheme of the CLC technology with solid fuels is shown in Fig. 1. In-situ gasification of solid fuel takes place into the fuel reactor, as well as

subsequent oxidation of the gasification products generated by gas-solid reactions with the oxygen carrier (M_xO_y). Thus, coal is oxidized to CO₂ and H₂O while the oxygen carrier is reduced. CO₂ in the product stream can be easily captured, once the water has been condensed. The reduced oxygen carrier (M_xO_{y-1}) is sent to the air reactor where it is oxidized with the oxygen in the air being ready to start a new redox cycle. In iG-CLC, the gasification is a slow step during the coal conversion, causing that some unconverted char could arrive at the air reactor, where it will be oxidized to CO₂, which would decrease the CO₂ capture efficiency of the process. A carbon separation step (e.g. carbon stripper reactor) between both fuel and air reactors is necessary to avoid this fact.

Complete combustion in case of gaseous fuels has been reached with the state of the art development of CLC [2]. However, complete combustion of coal to CO₂ and H₂O is not achieved so far in the iG-CLC process with solid fuels [3]. An oxygen polishing step would be required downstream the fuel reactor to oxidize unburnt products, which would require pure oxygen in order to maintain the advantage of the inherent CO₂ capture of the CLC process. Some technological solutions

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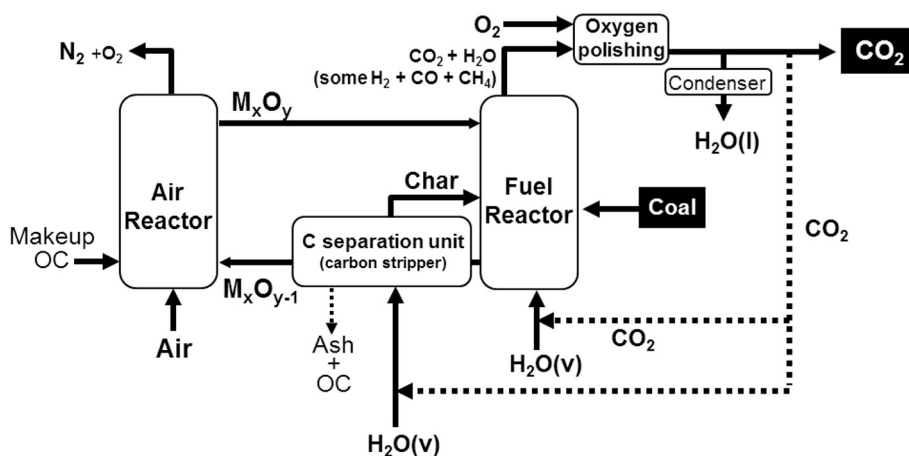


Fig. 1. Scheme of Chemical Looping Combustion with solid fuels.

have been proposed and evaluated in order to decrease the oxygen demand of the process [5]. In addition, the use of highly reactive oxygen carriers can help to decrease the oxygen demand of the iG-CLC process [6].

Ash produced during solid fuel combustion should be drained to avoid their accumulation in the system. In this case, oxygen carrier particles will be also extracted in the drainage, which causes a loss of oxygen carrier. A make-up stream of oxygen carrier will be then necessary. Similarly to the development of oxygen carriers for gaseous fuels, some synthetic materials have been evaluated as oxygen carrier for CLC of coal [7]. However, the relatively lower price of natural minerals or waste materials compared to synthetic materials, makes that these low-cost oxygen carriers are attractive for the application in iG-CLC technology. Ilmenite and iron ores have been fully investigated so far [3], as well as waste materials with high iron or manganese content [8–10]. More recently, Mn-ores are being considered as an alternative for Fe-based materials due to its relatively high reactivity and its positive effect on the char conversion rate. Several Mn-ores from different origin -Norway in [9,10], Gabon in [9,11], Colombia in [12], in addition to S. Africa, Brazil, Slovakia, Egypt in [13,14] or China and Ukraine in [15] - have been evaluated in thermogravimetric analysers or small fluidized bed facilities. Among them, Mn-ore from Brazil, Australia and Gabon have been considered as promising materials due to its high reactivity and good fluidization properties [11,16]. The char gasification rate in presence of these ores was faster than that observed in presence of other Fe-based materials such as ilmenite [17,18]. Alkali compounds (Na or K) transference from the Mn-ore to the char particles justified the improvement observed in char gasification [19]. The effect is similar to those observed with an iron material modified by K [20]. However, this effect was limited to a few redox cycles as Na or K was lost from the oxygen carrier material [18].

During 10 h of combustion with the Brazilian Mn-ore in a 10 kW_{th} CLC unit, improved CO₂ capture rates were attained compared to the use of ilmenite due to the enhancement of the char gasification rate [21]. The high attrition rate of this Brazilian ore forced to add new material continuously, thus incorporating fresh alkali elements, which are present in the Mn-ore, to the CLC unit. Similar conclusions were extracted from 18 h of combustion with ilmenite/Mn-ore mixtures and other Mn-ores in the 10 kW_{th} CLC unit [22,23], as well as with an Australian Mn-ore in a 100 kW_{th} CLC unit [24]. But the contrary was found during the combustion of biomass at the scale of 10 kW_{th} with braunite [25] and in a 2–4 MW_{th} unit with the Brazilian Mn-ore [26]. Nevertheless, the 2–4 MW_{th} unit is not optimized in terms of boiler temperature and oxygen carrier/fuel mixing for CLC operation, giving low combustion and CO₂ capture efficiency values.

Tested Mn-ores from Brazil and Australia showed high attrition rates, with estimated lifetime in the 100–400 h interval [11,23,24]

which is much lower than those estimated for an iron ore [27]. Recently, a Mn-ore from Gabon has been investigated in TGA and small fluidized bed reactor [11,18]. This material showed good fluidization properties, neither agglomeration of particles nor de-fluidization was observed, (even if the oxygen carrier was highly reduced) with a relatively low attrition rate, with an estimated particle lifetime of 1000 h. In addition, the enhancement of the char gasification rate in the presence of this Mn-ore was observed even when alkali compounds were lost after several redox cycles at high temperature. Therefore, this Mn-ore shows a great potential to be considered as oxygen carrier in CLC processes using solid fuels, but its behaviour during continuous coal combustion in a CLC unit must be still evaluated.

This work analyses the behaviour of the Mn ore from Gabon during 100 h in a 0.5 kW_{th} CLC unit for coal combustion. An extensive evaluation of the iG-CLC process was done, analysing the effect of main operating variables -such as temperature, oxygen carrier circulation rate, solids inventory and fluidizing medium- on the combustion efficiency and CO₂ capture. Results are further used to assess the potential of this Mn-ore to be used as oxygen carrier in comparison with the state of the art of Fe-based materials.

2. Experimental

2.1. Materials

A manganese ore from Gabon was used as oxygen carrier in the iG-CLC experiments. This material was supplied by Hidro Nitro Española S.A. Once received, it was crushed and sieved to the desired size (+100–300 μm). Then, particles of the manganese ore were thermally treated in air at 800 °C for 2 h to ensure the complete oxidation prior its utilization. According to previous works [28–30], particles in the +100–300 μm interval show good fluidization properties to be used in the 0.5 kW_{th} CLC unit at ICB-CSIC, where the fuel reactor is fluidized in the bubbling regime. Table 1 shows the main properties of this oxygen carrier. After calcination in air, the Mn-ore was mainly constituted by Mn₂O₃ and Fe₂O₃. However, Mn₂O₃ was easily reduced to MnO under combustion conditions, which mostly could be oxidized to Mn₃O₄ [11]. Thus, for cycled particles the oxygen transport capacity, R_{OC} , was calculated considering the Mn₃O₄-MnO redox pair, together the oxygen available in iron oxide considering the Fe₂O₃-Fe₃O₄ redox pair [31].

A South African bituminous coal with medium volatile matter content was fed into the fuel reactor during the experimental campaign. The diameter of the coal particles was in the +200–300 μm interval, which was suitable to minimize the elutriation of fuel particles from the fuel reactor [28]. The proximate and ultimate analyses of the coal are shown in Table 2. The lower heating value (LHV) of the South African coal is 26,430 kJ/kg.

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