



13th International Conference on Greenhouse Gas Control Technologies, GHGT-13, 14-18
November 2016, Lausanne, Switzerland

Performance of industrial residues as low cost oxygen carriers

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Abstract

In Chemical-looping combustion (CLC) an oxygen carrier is circulating, and transporting oxygen, between an air and fuel reactor, thus separating the flue gas stream from the nitrogen in the air. Except for the oxygen carrier all parts of a CLC system are conventional fluidized bed technology. Here, four iron-based industrial residues, and the conventional oxygen carrier ilmenite, are examined as oxygen carrier. The results indicate that two of the material, exhibit a better performance than ilmenite. This opens the possibility of not using virgin material as oxygen carrier and offers a new possible use for industrial residue material.

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Peer-review under responsibility of the organizing committee of GHGT-13.

Keywords: Chemical-looping combustion (CLC); oxygen carrier; industrial residue material

1. Introduction

Chemical-looping combustion (CLC) is a promising technology for burning fossil fuels with inherent CO₂ capture [1]. The general chemical-looping principle is shown in Fig. 1. CLC is generally carried out in two reaction steps. The fuel is used to reduce a solid oxygen carrier, typically a metal oxide, a reaction referred to as reduction. The reduced oxygen carrier is then reoxidized by air in a separate reaction step, thus referred to as oxidation. These two steps are commonly carried out in two separate reactors, the fuel reactor and the air reactor, with oxygen carrier's circulating

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between them. The gases from both reactors never mix and a stream of H₂O and CO₂ is obtained at the outlet of the fuel reactor which inherently stays separated from the air nitrogen.

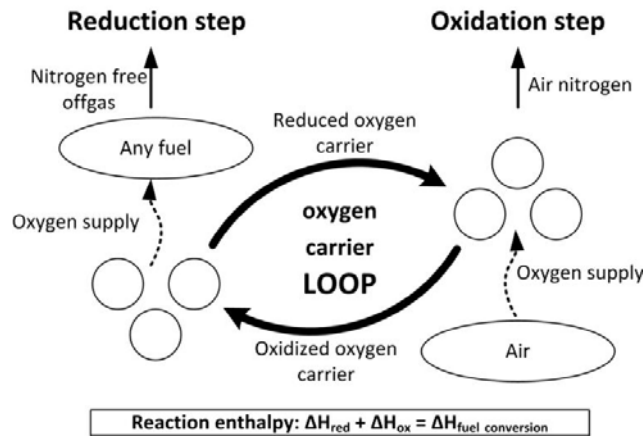


Fig. 1. Chemical-looping principle for thermochemical fuel conversion [2].

The oxidation of the reduced oxygen carrier in the air reactor is exothermic, whereas the reaction in the fuel reactor can be either exothermic or endothermic, depending on the choice of fuel and oxygen carrier. Overall the amount of heat produced is the same as for conventional combustion.

The principle of using solid oxygen carrier material to provide oxygen for thermochemical fuel conversion was proposed in the 1940s [3] to reform methane. The idea of CLC was patented as process for CO₂ production from carbonaceous sources in the 50s [4]. The same principle was proposed later on with the goal to lower the irreversibility of combustion [5, 6]. With the increasing recognition of anthropogenic climate change, the concept was investigated to recover CO₂ from the exhaust of combustion systems for power generation [7, 8]. Since then, obtaining CO₂ capture with low energy penalty for power sector is the main driving force for the CLC research.

Gaseous fuels [9-11], such as natural gas or syngas, and solid fuels [12-18], such as coal or biomass, have been widely investigated in the literature as possible fuels in CLC. While the application of gaseous fuels appears to be less problematic, for solid fuels, losses of oxygen carriers due to possible side reaction with ash [19], as well as material withdrawn from the reactor with the ash, may lead to high expected amounts of oxygen carrier makeup [18]. This increases the demand for abundant, low cost and environmental friendly oxygen carrier materials [14]. Possible materials under investigation have been natural ores [20-24] and industrial residues or by-products [25-28]. Among the natural ores, ilmenite has been most widely investigated [21, 29-32]. Five kinds of ilmenite from different resources was compared (Norway, Canada, South Africa and Madagascar) [23] and the reactivity of the Norwegian ilmenite was higher than the others. It was found that the reactivity of fresh materials increases with successive redox cycles. This is referred to as activation [33].

Apart from using ores and industrial residues in CLC solely for the purpose of electric power generation, CLC with these materials could also be integrated into other industrial processes. The extractive industry, as well as the steel industry, generates large CO₂ emissions. If one could find promising oxygen carrier materials in the related processes, CLC could be introduced into the process chain to lower the carbon footprint of these industries, e.g. for hematite or ilmenite a combination of CLC with a smelting process is proposed in a German patent [34]. A screening of Fe-based ores and by-products found that two samples, SSAB Röd and SSAB Brun, showed good and stable reactivity [26]. But these materials came in the form of very fine dust and the required production of particles would therefore increase the cost of the carriers. Iron oxide scale [26, 28] was also investigated as oxygen carriers but its reactivity was unstable and rather low.

The conversion of solid fuels in the fuel reactor consists of three steps - pyrolysis, gasification and combustion. Pyrolysis of the fuel takes place immediately after the fuel is introduced into the fuel reactor, producing volatiles and

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