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# Study of the performances of an oxygen carrier: Experimental investigation of the binder's contribution and characterization of its structural modifications



*Performances d'un porteur d'oxygène : étude expérimentale de la contribution du support et caractérisation de ses modifications structurales*

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## ABSTRACT

The aim of this work is to investigate the contribution of the binder (NiAl<sub>2</sub>O<sub>4</sub>) on the performances of the oxygen carrier NiO/NiAl<sub>2</sub>O<sub>4</sub>. To this purpose, oxidation/reduction cycles have been performed in a fixed bed reactor using CO as a fuel. The results reveal that the binder can react with the fuel to form CO<sub>2</sub>, and that its total reduction capacity increases with temperature. XRD characterizations performed on the binder (on the fresh and after several cycles) show a shift of the diffraction peaks of NiAl<sub>2</sub>O<sub>4</sub> toward the ones of γ-alumina, which can be attributed to a progressive decomposition of NiAl<sub>2</sub>O<sub>4</sub> to alumina and NiO.

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## R É S U M É

L'objectif de ce travail est d'étudier la contribution du liant (NiAl<sub>2</sub>O<sub>4</sub>) sur les performances du transporteur d'oxygène NiO/NiAl<sub>2</sub>O<sub>4</sub>. Des cycles d'oxydation/réduction ont ainsi été réalisés dans un réacteur en lit fixe, avec CO comme combustible. Les résultats montrent que le liant réagit avec le combustible pour former CO<sub>2</sub>, et que sa capacité totale de réduction augmente avec la température. Des études de DRX réalisées sur le liant (sur le matériau frais et après quelques cycles) révèlent un déplacement des pics de NiAl<sub>2</sub>O<sub>4</sub> vers ceux de l'alumine gamma, qui peut être attribué à la décomposition de NiAl<sub>2</sub>O<sub>4</sub> en Al<sub>2</sub>O<sub>3</sub> et NiO.

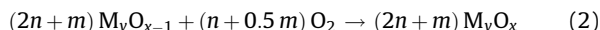
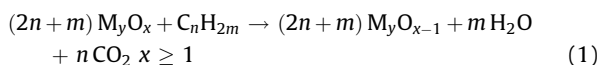
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## 1. Introduction

The increasing concentration of greenhouse gases in the atmosphere contributes to global warming and climate change. CO<sub>2</sub> is the main anthropogenic greenhouse gas and its concentration in the atmosphere has increased from a preindustrial value of about 280 ppm to reach 401 ppm (monthly averages) in April 2014 [1]. It is generally accepted that CO<sub>2</sub> emissions should be significantly reduced, as a preventive action and in order to stabilize its concentration in the atmosphere, to limit its environmental impact. In 2010, 41% of the worldwide anthropogenic CO<sub>2</sub> emissions came from fossil fuel (coal, oil or natural gas) combustion to produce energy [2]. Since transition to renewable energies is a slow process and fossil fuel will still be the dominant energy source in the next decades, CO<sub>2</sub> capture and sequestration (CCS) has been proposed as an alternative to reduce CO<sub>2</sub> emissions from fossil fuel power plants. CCS is a process consisting in the separation of CO<sub>2</sub> from industrial and energy-related flue gases, transport to a storage location and long-term isolation from the atmosphere [3]. The current CO<sub>2</sub> capture systems consist in pre-combustion, oxy-fuel combustion and post-combustion systems. These techniques have a main disadvantage; they exhibit a high-energy penalty, which decreases the overall efficiency of the power plant by around 15–20% [4]. Many efforts are undertaken to develop new inexpensive CCS technologies [5]. The chemical looping combustion (CLC) has proven to be an efficient alternative to reduce the cost of CO<sub>2</sub> capture. It is an innovative technology, which allows the production of energy with a selective capture of CO<sub>2</sub> and low energy penalty [6,7]. In this cyclic process, the oxygen needed for the combustion is provided by an oxygen carrier that is alternatively reduced by the fuel according to reaction (1) and oxidized by air (reaction (2)).



The main advantage of this technology compared to conventional combustion is that the combustion is carried out without air. Thus, exhaust gases from the fuel combustion reaction are mainly composed of CO<sub>2</sub>, H<sub>2</sub>O; after water condensation, pure CO<sub>2</sub> can be obtained. The gas mixture produced during re-oxidation of the oxygen carrier contains nitrogen and some unreacted oxygen. So, it can be directly released to the atmosphere. Moreover, this process also ensures that no thermal NO<sub>x</sub> are produced in the flue gas, due to the absence of extremely high temperatures [8]. Reaction (2) is extremely exothermic, while reaction (1) can be endothermic or exothermic, depending on the metal oxide and the fuel used. The heat balance during the CLC process is the same as the one released during conventional combustion.

This cyclic process can be carried out in two ways: by moving the oxygen carrier between two static gas flows or by maintaining the oxygen carrier static while switching the gas streams. The first option is carried out in two interconnected fluidized bed reactors, whereby the oxygen carrier is continuously circulated between a fuel reactor, where it is reduced and an air reactor where it is regenerated [9,10] (see Fig. 1a). This option has been widely studied since the 1980s and several units for gaseous fuels can be found in the literature (from 10 kW<sub>th</sub> units [11–13] to 1 MW [14]). The second option (see Fig. 1b) was first proposed by Noorman et al. [15,16] and consists in using dynamically operated packed-bed reactors where the particles remain stationary and a valve system switches the gas flow periodically. In this case, the difficult gas-solid separation is avoided.

The success of a large-scale CLC application depends on finding suitable oxygen carriers that exhibit significant oxygen transport capacity, fast reduction/oxidation rates and chemical/physical stability [17]. Several metal oxides such as NiO [18], CuO [19,20], Mn<sub>3</sub>O<sub>4</sub> [21,22] and Fe<sub>2</sub>O<sub>3</sub> [23,24] have been proposed as potential candidates for CLC. These metal oxides cannot be used alone, as their rate

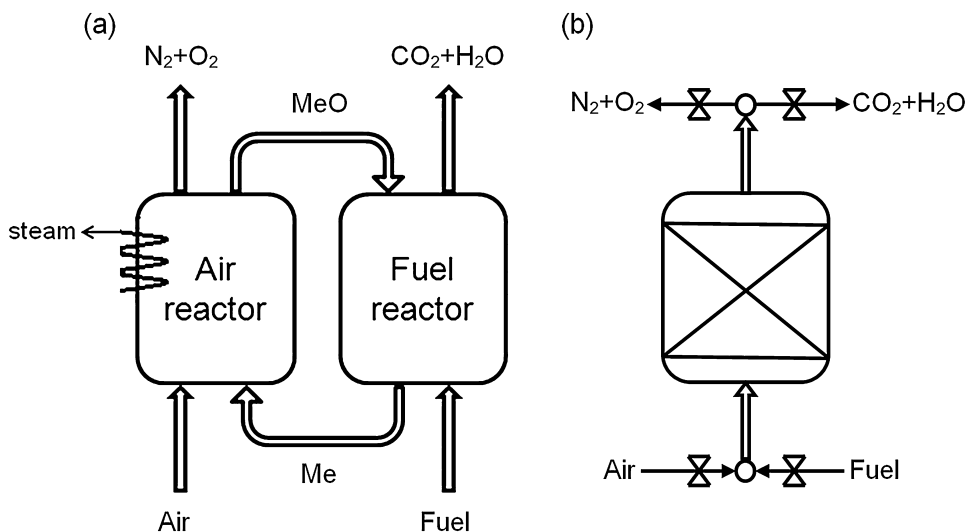


Fig. 1. Possible CLC configurations: (a) interconnected fluidized beds, (b) packed-bed.

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