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Experimental investigation of some metal oxides for chemical looping combustion in a fluidized bed reactor

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ABSTRACT

Chemical looping combustion (CLC) is the process in which metal oxides, rather than air or pure oxygen, supply the oxygen required for combustion. In this process, different gaseous fuels can be burnt with the inherent separation of CO_2. The feasibility of the CLC system depends greatly on the selection of appropriate metal oxides as oxygen carriers (OC). In this study, NiO–$NiAl_2O_4$, $Cu_{0.95}Fe_{1.05}AlO_4$, and CuO–$Cu_{0.95}Fe_{1.05}AlO_4$ were tested experimentally in a fluidized bed reactor as a function of oxidation–reduction cycles, temperature, bed inventory and superficial gas velocity. The results showed that flue gases with a CO_2 concentration as high as 97% can be obtained. The flue gases should be suitable for transport and storage after clean-up and purification. With an increase in the bed inventory or a decrease in superficial gas velocity, the flue gas characteristics improved i.e. more CO_2 and fewer secondary components or less unreacted fuel were obtained. Carbon formation could occur during the reduction phase but it decreased with an increase in temperature and inventory and could be completely avoided by mixing steam with the fuel. The reactivity of NiO/$NiAl_2O_4$ was higher than the Cu- and Fe-based oxygen carriers. Increasing the CuO fraction in the oxygen carrier led to defluidization of the bed during the reduction and oxidation phases.

1. Introduction

CO_2 is one of the major anthropogenic greenhouse gases emitted into the atmosphere. One solution is to capture CO_2 and store it permanently and safely under geological formations or deep in the sea. Chemical Looping Combustion (CLC) can be used for clean power generation as it may have a lower energy penalty due to its inherent separation of CO_2 [1] and may also be a better choice for CO_2 capture. During the last decade, considerable research has been carried out on the use of CLC for the combustion of gaseous fuels [2–9].

Chemical Looping Combustion (CLC) is an oxy-combustion in which the oxygen required for combustion is supplied by metal oxides known as oxygen carriers (OC). The CLC concept, as shown in Fig. 1, consists of two interconnected reactors known as the air and the fuel reactor. The oxygen carriers are reduced by the gaseous fuel in the fuel reactor. The reduced oxygen carriers are transferred to the air reactor where they are oxidized by air. The reactions in the air reactor are exothermic, and exothermic or endothermic in the fuel reactor depending upon the fuel and the oxygen carriers. The flue gases leaving the air reactor contain N_2 and the unreacted O_2 of air. Flue gases from the fuel reactor contain mostly CO_2, H_2O, a fraction of CO, H_2, if there is partial conversion of the fuel. However, flue gases from the fuel reactor do not contain N_2 from the air, which is the major component of flue gases from the classical combustion techniques. The partial pressure of CO_2 in the flue gases from the fuel reactor is very high. For fuels containing only C and H, pure CO_2 can be obtained after H_2O condensation.

The success of the CLC system is dependent on finding suitable oxygen carriers that have sufficiently high conversion rates in cyclic conditions of oxidation and reduction, high communition and agglomeration resistance, and are economical and environmentally friendly. The metal oxides of Ni, Cu, Fe, Co, and Mn are being tested by different researchers at different scales [10–13]. Various support materials, namely Al_2O_3, $MgAl_2O_4$, ZrO_2, and TiO_2 are used to enhance some of the desirable characteristics of oxygen carrier particles.

Ni-based oxygen carriers are considered among the most suitable for CLC. Research on these compounds has shown that YSZ, Al_2O_3, $NiAl_2O_4$, SiO_2, bentonite, and NiO/hexaaluminate can be used as a support material for NiO [14,15], while Jin et al. [16]

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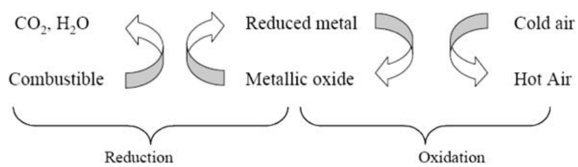

Fig. 1. Principle of chemical looping combustion.

consider that $NiAl_2O_4$ is one of the best supports. Cu- and Fe-based oxygen carriers are also good candidates. However, Cu-based oxygen carriers are limited to a lower operating temperature due to the low melting point of Cu. Fe-based OC are associated with a low oxygen-carrying capacity. Recently, a new type of material has been developed that contains oxides of copper and iron and uses Al_2O_3 as a support.

In this study, reduction–oxidation cycles under different conditions of fluidization were analyzed. The objective of this work was to study the performances of newly developed Cu–Fe-based oxygen carriers and $NiO/NiAl_2O_4$ under different experimental operating conditions: temperature, bed inventory and gas superficial velocity in the fluidized bed reactor.

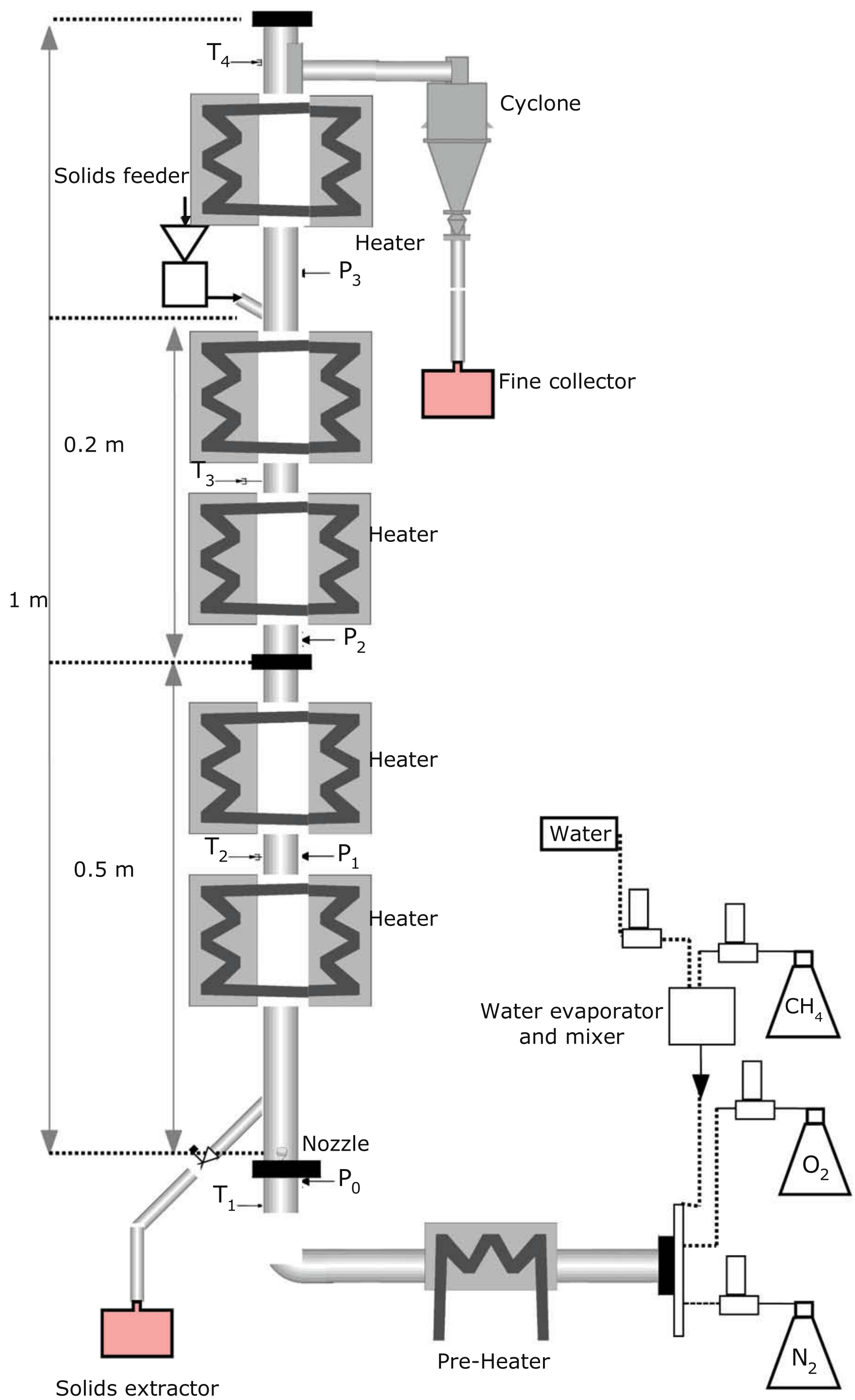

Fig. 2. Experimental equipment.