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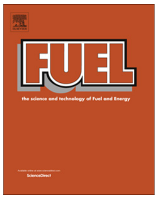


Design of a rotary reactor for chemical-looping combustion. Part 2: Comparison of copper-, nickel-, and iron-based oxygen carriers

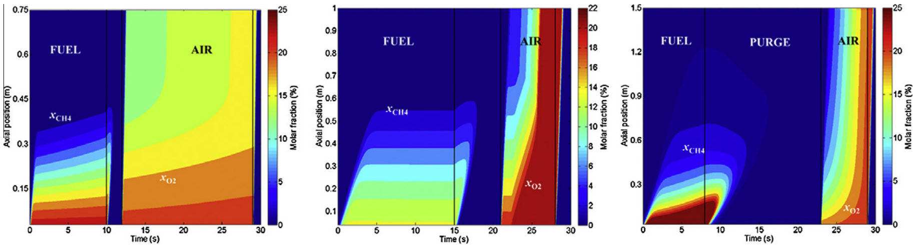
Zhenlong Zhao, Ahmed F. Ghoniem *

Department of Mechanical Engineering, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, MA 02139-4307, USA

HIGHLIGHTS

- The rotary CLC designs have been proposed with Cu-, Ni-, and Fe-based OCs.
- For all OCs, complete fuel conversion and carbon separation can be achieved.
- Ni-based design is limited by oxidation; Cu- and iron-based cases are limited by reduction.
- Reduction kinetics, pressure, and temperature are the most important parameters.
- The design can be readily scaled to different conditions with desirable performances.

GRAPHICAL ABSTRACT



Nickel Copper Iron

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ABSTRACT

Chemical-looping combustion (CLC) is a novel and promising option for several applications including carbon capture (CC), fuel reforming, H_2 generation, etc. Previous studies demonstrated the feasibility of performing CLC in a novel rotary design with micro-channel structures. Part 1 of this series studied the fundamentals of the reactor design and proposed a comprehensive design procedure, enabling a systematic methodology of designing and evaluating the rotary CLC reactor with different OCs and operating conditions. This paper presents the application of the methodology to the designs with three commonly used OCs, i.e., copper, nickel, and iron. The physical properties and the reactivities of the three OCs are compared at operating conditions suitable for the rotary CLC. Nickel has the highest reduction rate, but relatively slow oxidation reactivity while the iron reduction rate is most sensitive to the fuel concentration. The design parameters and the operating conditions for the three OCs are selected, following the strategies proposed in Part 1, and the performances are evaluated using a one-dimensional plug-flow model developed previously. The simulations show that for all OCs, complete fuel conversion and high carbon separation efficiency can be achieved at periodic stationary state with reasonable operational stabilities. The nickel-based design includes the smallest dimensions because of its fast reduction rate. The operation of nickel case is mainly limited to the slow oxidation rate, and hence a relatively large share of air sector is used. The iron-based design has the largest size, due to its slow reduction reactivity near the exit or in the fuel purge sector where the fuel concentration is low. The gas flow temperature increases monotonically for all the cases, and is mainly determined by the solid temperature. In the periodic state, the local temperature variation is within 40 K and the thermal distortion is limited. The design of the rotary CLC is also scaled to different pressures and inlet temperatures. The method of scaling is discussed and desirable operational performances are obtained.

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* Corresponding author. Tel.: +1 617 253 2295.
E-mail address: ghoniem@mit.edu (A.F. Ghoniem).

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Nomenclature

Symbols

A	cross-sectional area, m^2
a	pressure coefficient
C_i	concentration of species i, mol m^{-3}
D	reactor diameter, m
d	channel width, m
E_a	activation energy, J mol^{-1}
E_g, E_s	energy of gas or solid, J m^{-3}
H	channel height, m
H_g	enthalpy of gas, J m^{-3}
h_{gs}	heat transfer coefficient between phases, W m^{-2} K^{-1}
$h_{m,i}$	external mass transfer coefficient, m s^{-1}
$\hat{h}_{s,i}$	molar enthalpy for species, J mol^{-1}
k, k_p	reaction rate constant, $m^{(3n-3)}$ $mol^{(1-n)}$ s^{-1}, n is the reaction order
k_s, k_g	thermal conductivity of solid or gas phase, W m^{-1} K^{-1}
k_0	pre-exponential factor, $m^{(3n-3)}$ $mol^{(1-n)}$ s^{-1}, n is the reaction order
m_{ox}	mass of fully oxidized oxygen carrier, kg
m_{red}	mass of fully reduced oxygen carrier, kg
n	reaction order
P	operating pressure, Pa
P_c	inner perimeter of the channel, m
$p_{i,out}$	partial pressure of species i, Pa
Q_{gs}	heat flux from gas phase to solid phase, W m^{-2}
T	temperature, K
u	velocity, m s^{-1}
X	conversion of oxygen carrier
x_i	molar fraction of species i

Greek letters

δ_{bulk}	thickness of the bulk support layer, m
δ_{oc}	thickness of the porous oxygen carrier layer, m
δ_s	thickness of the solid phase (including the porous layer and the bulk layer), m
ε_i	volume fraction of phase (or species) i
ε_M	cross-section area ratio of solid
ε_g	porosity of the solid
ν	stoichiometric coefficient
θ_i	size of sector i, rad
ρ	density, kg m^{-3}
$\hat{\rho}$	molar density, mol m^{-3}
τ	cycle period, s
ω	overall molar reaction rate, mol m^{-2} s^{-1}

Acronyms

CC	carbon capture
CLC	chemical-looping combustion
OC	oxygen carrier
Redox	reduction and oxidation

1. Introduction

Chemical-looping combustion (CLC) is a novel and promising technology for carbon capture (CC). In CLC, the combustion process is performed in two reactors: a fuel reactor and an air reactor. A solid oxygen carrier (OC) is circulated between these two reactors to transport undiluted oxygen from air to fuel. The exhaust gas from reduction contains only CO_2 and steam, and pure CO_2 can be readily obtained after water condensation. Using OCs as the looping medium, the direct contact between air and fuel is circumvented and hence energy-intensive gas separation processes are avoided.

CLC is mostly carried out in two interconnected fluidized-bed reactors with the OC in the form of particles circulating in between [1–7]. Alternatively, a rotary reactor consisting of a large number of micro-channels was proposed in previous studies [8–12]. As shown in Fig. 1a, the rotary reactor has a rotary wheel, which rotates through four sectors, i.e., fuel, air, and two steam purging sectors. The wheel consists of a large number of channels (Fig. 1b) with the OC coated onto their inner walls. As shown in Fig. 1c, the channel wall has two solid layers: a highly porous OC layer and a bulk dense inert substrate. The porous layer consists of active metal oxides, as well as inert binders to maintain the pore structures. Pressurized streams flow though the reactor and react with the OCs to generate the product stream, in this case CO_2/H_2O, and the oxygen-depleted air stream. A complete description of the reactor design and functionality can be found in Refs. [12,13].

Few studies have been carried out to examine the performances of the rotary reactor for CLC under different conditions [8,9,11,14]. Pavone and co-workers [8,9] simulated the initial reduction and oxidation (redox) cycles of the reactor with NiO/Al_2O_3 as the OC, and observed 90% separation efficiency but large temperature variations. Zhao et al. [11] examined a copper-based design with a dense boron-nitride support layer, and obtained a stable and periodically stationary performance with complete fuel conversion and carbon separation. However, the behavior strongly depends on the choice of the OCs, the designs, and the operating conditions [14]. From these studies, it is clear that the design of a rotary CLC reactor is a complex process: many parameters are closely involved and coupled, all of which strongly affect the functionalities of the reactor. Thus, a comprehensive parametric study is important to understand the intrinsic logics and develop the optimized procedures of design under different conditions.

The objective of this two-part series is to investigate the fundamental relations among the design parameters, develop a systematic design procedure, and compare the performances of the design under different conditions. In Part 1, the fundamental effects of the OC characteristics, the reactor configurations, and the operating conditions are examined. A systematic procedure is proposed on the basis of the parametric study. Part 2 presents the application of this procedure to three commonly used OCs, i.e., copper, nickel, and iron. The reaction mechanisms utilized in this study are based on the one-step overall kinetics proposed by Abad and co-workers [15–17]. The redox reactivities of the OCs are analyzed and compared, and their effects on the choice of design are discussed. The performances of rotary reactor with the three OCs are illustrated and the key parameters for each case are identified. The effects of the operating temperature and pressure on the design are also studied and the scaling strategies to different conditions are proposed.

2. Reactor design

A number of criteria should be satisfied for the design of a rotary reactor, including complete (>99%) fuel conversion, sufficient (>95%) CO_2 separation, and adequate operational stabilities. To achieve these objectives, three groups of parameters can be specified, (i.e., the material selection, the reactor configuration, and the operational conditions), following the procedure proposed in Part 1 (see Fig. 7 in Ref. [13]). For the scope of this study, the design