



Design of a rotary reactor for chemical-looping combustion. Part 1: Fundamentals and design methodology



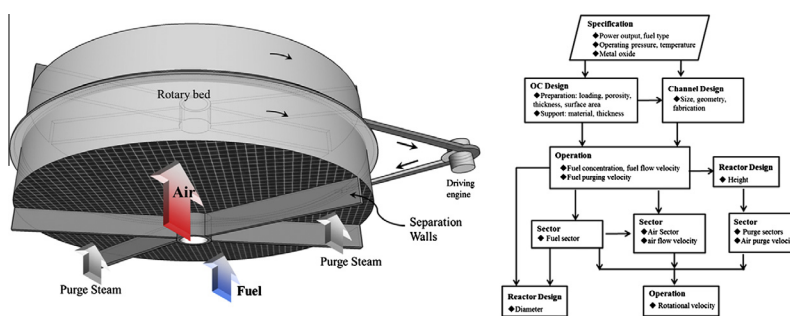
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HIGHLIGHTS

- A novel rotary reactor has been proposed for CLC with carbon separation.
- The design is compact, stable, and it captures CO₂ with minimum energy penalty.
- The fundamentals of OC, reactor design, and operating conditions have been studied.
- The key parameters for the reaction kinetics have been evaluated by simple models.
- A systematic procedure has been proposed for reactor design and optimization.

GRAPHICAL ABSTRACT



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ABSTRACT

Chemical-looping combustion (CLC) is a novel and promising option for several applications including carbon capture (CC), fuel reforming, H₂ generation, etc. Previous studies demonstrated the feasibility of performing CLC in a novel rotary design with micro-channel structures. In the reactor, a solid wheel rotates between the fuel and air streams at the reactor inlet, and depleted air and product streams at exit. The rotary wheel consists of a large number of micro-channels with oxygen carriers (OC) coated on the inner surface of the channel walls. In the CC application, the OC oxidizes the fuel while the channel is in the fuel zone to generate undiluted CO₂, and is regenerated while the channel is in the air zone. In this two-part series, the effect of the reactor design parameters is evaluated and its performance with different OCs is compared. In Part 1, the design objectives and criteria are specified and the key parameters controlling the reactor performance are identified. The fundamental effects of the OC characteristics, the design parameters, and the operating conditions are studied. The design procedures are presented on the basis of the relative importance of each parameter, enabling a systematic methodology of selecting the design parameters and the operating conditions with different OCs. Part 2 presents the application of the methodology to the designs with the three commonly used OCs, i.e., nickel, copper, and iron, and compares the simulated performances of the designs.

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1. Introduction

It has been widely accepted that fossil fuel combustion is a major contributor to the rise of CO₂ concentration in the

atmosphere and global warming. One approach to reduce anthropogenic CO₂ emissions, apart from improving energy efficiency and using alternative sustainable energy sources, is carbon capture and sequestration (CCS), in which CO₂ is separated from flue gases, liquefied, and injected in geological formations, such as depleted oil or gas fields. Of these steps, CO₂ capture remains the most challenging part from an economic point of view since obtaining

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Nomenclature

Symbols

A	cross-sectional area, m^2
C_i	concentration of species i , mol m^{-3}
C_f	friction coefficient
c	specific heat capacity, $\text{J kg}^{-1} \text{K}^{-1}$
D	reactor diameter, m
d_h	channel width, m
H	channel height, m
h_{gs}	heat transfer coefficient between phases, $\text{W m}^{-2} \text{K}^{-1}$
h_m	external mass transfer coefficient, m s^{-1}
Δh_r	enthalpy of reaction, J mol^{-1}
k	reaction rate constant
k_s, k_g	thermal conductivity of solid or gas phase, $\text{W m}^{-1} \text{K}^{-1}$
k_0	pre-exponential factor
m_{ox}	mass of fully oxidized oxygen carrier, kg
m_{red}	mass of fully reduced oxygen carrier, kg
\dot{N}_f	molar flow rate of fuel
P	operating pressure, Pa
P_c	inner perimeter of the channel, m
p_i	partial pressure of species i , Pa
R_{oc}	oxygen transport capacity
R_u	gas universal constant, $8.314 \text{ J mol}^{-1} \text{K}^{-1}$
T	temperature, K
t	gas residence time in the channel, s^{-1}
u	velocity, m s^{-1}
\dot{W}_{th}	target thermal capacity, W

X	conversion of oxygen carrier
x_i	molar fraction of species i
$z_{99\%}$	height to reach 99% fuel conversion, m

Greek letters

δ_{bulk}	thickness of the bulk support layer, m
δ_{oc}	thickness of the porous oxygen carrier layer, m
ε	porosity of the OC
η_i	fuel conversion efficiency, percentage of fuel converted in the reactor
γ_{CH_4}	conversion yield of CH_4
κ	redundancy factor
μ_g	viscosity of gas, $\text{kg m}^{-1} \text{s}^{-1}$
ν	stoichiometric coefficient
θ_i	size of sector i , rad
ρ	density, kg m^{-3}
τ	cycle period, characteristic time of reaction, channel residence time, s
ω	rotational velocity, s^{-1}

Acronyms

CC	carbon capture
CCS	carbon capture and sequestration
CLC	chemical-looping combustion
OC	oxygen carrier
Redox	reduction and oxidation
TIT	turbine inlet temperature

carbon dioxide in high purity still accounts for the major share of the cost of state-of-the-art CCS technologies. In the past few years, extensive research focus has been placed on three general processes for capturing CO_2 from combustion in power plants: post-combustion capture, pre-combustion decarbonization and oxy-combustion. One of the key issues that limits the applications of CCS approaches is the large energy penalty during the separation process.

Recently, a new approach for CO_2 capture has been widely investigated. This approach was named “chemical-looping combustion (CLC)” [1] and belongs to oxy-fuel combustion. In CLC, combustion is decomposed into two steps: fuel is oxidized by a metal oxide in a fuel reactor to generate CO_2 and steam; the reduced metal oxide is then regenerated by air in an air reactor. During this two-step process, the looping medium acts as an “oxygen carrier” (OC), which adsorbs oxygen in the air reactor and releases it to oxidize fuel in the fuel reactor. The main advantage of CLC is that by using OCs as the medium to transport the pure oxygen, the direct contact between air and fuel is circumvented and hence energy-intensive gas separation processes are avoided.

CLC is commonly carried out by physically transporting the OC particles between two interconnected fluidized bed reactors [2–4]. In this reactor concept, the OCs in the form of particles are fluidized and pneumatically transported continuously between the fuel and air reactors. Several CLC units using the fluidized-bed design have been built and operated by Chalmers University of Technology of Sweden [5–8], ICB-CSIC of Spain [9–11], Vienna University of Technology of Austria [12,13], and Southeast University of China [14,15]. Meanwhile, some novel reactor designs have emerged such as the moving-bed reactor in Ohio State University [16,17], the fixed packed-bed reactor in Eindhoven University of Technology of Netherlands [18,19], and the rotating packed-bed reactor in SINTEF Materials & Chemistry of Norway [20,21].

Recently, a rotary reactor design with micro-channel structures was proposed [22,23]. As shown in Fig. 1a, the reactor

consists of a rotary wheel and two stationary chambers at the top and bottom of the wheel. The wheel rotates continuously through four sectors (Fig. 1b): fuel, air, and two purging sectors. The rotary wheel consists of a large number of micro-channels (Fig. 1c) with the OC coated onto their inner wall. As shown in Fig. 1d, the channel wall has two solid layers with one being a highly porous OC layer and the other being a bulk dense ceramic layer with high thermal inertia and thermal conductivity. The OC layer usually consists of an active metal oxide which reacts with the fuel and air alternatively, as well as an inert ceramic substrate which helps maintain the pore structures and chemical reactivities of the OC. As seen in Fig. 2, pressurized feed gas (fuel, air or steam) flows through the reactor, reacts with the OC while it is heated to high temperature. In the fuel sector, the fuel is diluted with CO_2 to effectively lower the operating temperature while in the air sector air is used to re-oxidize the OC. Two purging sectors are implemented between the fuel and the air sectors to sweep the residual gases out of the reactor. Flue streams from a large number of channels merge into two separate streams from the fuel zone and the air zone (see Fig. 2b), respectively, and then drive turbines in the downstream, as described in Ref. [22]. The main incentives for performing CLC in this way are (1) the separation of the gas and particles is intrinsically avoided, (2) the operation is continuous and stationary, and (3) the design is compact and easy to scale.

Pavone and co-workers [24,25] simulated the reduction and oxidation (redox) performances of the rotary reactor with nickel oxide wash-coated on alumina substrate using COMSOL for the initial cycles. Because of the limited amount of the solid phase in the base-case design, large temperature fluctuations (500°C) were observed in the solid phase, which renders the design unstable over repeated cycles. Zhao et al. [26] investigated the periodic stationary-state performances of a rotary design with copper oxide supported on boron nitride substrate using a one-dimensional plug-flow model. The base-case simulation validated the

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