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Modeling and scale analysis of gaseous fuel reactors in chemical looping combustion systems

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ABSTRACT

This work investigates the scale-up of chemical looping combustion (CLC), a next-generation technology for carbon capture and storage, to the industrial scale. The study focused on the bottom bed of the unit, which was considered to be the critical region during scale-up due to the large solids inventory in this zone combined with relatively inefficient gas–solids contact. Two CLC reactors of vastly different sizes (bench and utility scale) were studied to discern their difference related to scale-up via a one-dimensional model. This model considered kinetics that varied with the degree of oxidation and population distribution of the oxygen carriers, the mixing of which accounts for both convective and dispersive transport. The model was validated against bench scale data, and was used to evaluate the performance of a 1000 MW_{th} CLC fuel reactor using either syngas or methane as fuels. Sensitivity analyses were also carried out with this model to determine the effects of several parameters on fuel conversion, including solids circulation, oxygen carrier reactivity, bed height, and maximum bubble size. The results show that the mass transfer of gas from bubbles to the emulsion phase represents a significant limiting factor for fuel conversion in the bottom bed of a utility scale fuel reactor.

Introduction

The Intergovernmental Panel on Climate Change (IPCC) has concluded that emissions of greenhouse gases, in particular CO_2, as the result of fossil fuel combustion have increased markedly during the past several centuries and have resulted in greater radiative forcing (IPCC, 2007). According to the IPCC (2014) the energy supply sector is the single largest source of global greenhouse gas emissions, and carbon capture and storage (CCS) is a key element in several possible scenarios for CO_2 mitigation. The principle of CCS is to capture and store the CO_2 generated from energy-intensive facilities, thus preventing its release to the atmosphere.

Chemical looping combustion (CLC) is a fuel conversion method with the potential to separate CO_2, and so is applicable to CCS. In this technology, a metal oxide (termed an oxygen carrier) circulates between two fluidized bed reactors. The concept was first suggested for power production purposes in 1994, using nickel as the oxygen carrier (Ishida & Jin, 1994). Since then, this technology has been widely researched, with a review by Adanez, Abad, Garcia-Labiano, Gayan, and de Diego (2012). In an air reactor (Fig. 1), which is fluidized by air, the oxygen carrier is interfaced with an oxidizing atmosphere. In contrast, in a fuel reactor, fluidized by steam and/or fuel gas, a reducing atmosphere (relative to the oxygen carrier) is present. The metal oxide is therefore reduced in the fuel reactor, providing oxygen for fuel oxidation (Eq. (1)), and subsequently re-oxidized in the air reactor (Eq. (2)).

$$C_nH_{2m} + (2n+m)\,Me_xO_y \rightarrow nCO_2 + mH_2O + (2n+m)\,Me_xO_{y-1} \quad (1)$$

$$O_2 + 2Me_xO_{y-1} \rightarrow 2Me_xO_y \quad (2)$$

It is important that the gas exiting the fuel reactor is oxidized to the greatest extent possible. Unreacted fuel would require additional combustion steps relying on oxy-fuel combustion or similar processes, impacting the cost and efficiency of the process.

The modeling of fluidized bed combustion has been developed over the last several decades, such as by Hannes (1996). The models reported in the literature can be divided into three distinct classes with respect to the degree of abstraction: modeling based on experimental correlations, semi-empirical modeling, and computational fluid dynamics (CFD). Correlation-based models are widely used by large-scale circulating fluidized bed (CFB) manufacturers and are based on experimental data acquired from existing equipment. These in-house correlations are typically unpublished and are used for interpolation (or even some level of extrapolation) to predict

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Nomenclature

A	Cross-sectional area, m^2
b	Stoichiometric coefficient, mol_{OC}/mol_{fuel}
C_{em}, C_{bub}	Gas concentrations in the emulsion and bubble phases, kg/m^3
C_s	Solids concentration, kg/m^3
D_j	Diffusion coefficient for compound j, m^2/s
$D_{s,z}$, $D_{g,z}$	Axial solids or gas dispersion coefficient, m^2/s
d	Diameter, m
d_B	Bubble diameter, m
E_A	Activation energy, J/mol
$f(\omega)$	Function for the effect of ω
f_{OF}	Stoichiometric ratio of oxygen consumed per mole of fuel oxidized
g	Gravitational acceleration, m/s^2
H_b	Bed height, m
H_{LHV}	Lower heating value on a molar basis, J/mol
$(K_{Be})_B$	Gas interchange coefficient, $m^3/(m^3\,s)$
k	Rate constant, $mol^{1-n}(m^3)^{n-1}/s$
k_0	Pre-exponential factor, $mol^{1-n}(m^3)^{n-1}/s$
k_{eff}	Mass-based effective reaction rate constant, s^{-1}
k_F	Flow-normalized, mass-based reaction rate constant, $Nm^3/(t\,s)$
M	Molar mass, kg/mol
m	Mass, kg
$\dot{m}_s$	Mass flow of solids, kg/s
N_g	Number of gas species
N_{OC}	Number of oxygen carrier classes
N_ψ	Number of reactions
n	Reaction order
$\dot{n}$	Molar flow, mol/s
p_j, p_{tot}	Partial pressure, total pressure, Pa
R	Universal gas constant
R_0	Oxygen carrier theoretical maximum oxygen transfer capacity
r_g	Grain radius, m
r_{hom}	Reaction rate for homogeneous reactions, $mol/(m^3\,s)$
r_ψ	Reaction rate for heterogeneous reaction ψ, s^{-1}
S	Source term, $kg/(m^3\,s)$
T	Temperature, K
u	Velocity, m/s
V_n	Normal molar volume, Nm^3/mol
X	Degree of conversion of oxygen carrier
Y	Molar fraction
z	Coordinate axis along the height of the unit, m

Greek letters

α	Oxygen consumed by reaction, $mol/(m^3\,s)$
β	Stoichiometric factor, mol_{fuel}/mol_O
γ	Degree of conversion
δ_B	Bubble fraction
ε_{mf}	Minimum fluidization voidage
λ	Stoichiometric oxygen ratio
ρ_p	Particle density, kg/m^3
ρ_s	Molar density of oxygen carrier, mol/m^3
ω	Degree of oxidation

Subscripts

0	Superficial
bub	Bubble
em	Emulsion
excess	Excess gas
g	Gas
het	Heterogeneous
hom	Homogeneous
i	Oxygen carrier class
j	Gas species
mf	Minimum fluidization
O	Oxygen
OC	Oxygen carrier
ox	Oxidized state
red	Reduced state
s	Solid
t	Terminal
tp	Two-phase
ψ	Reaction number

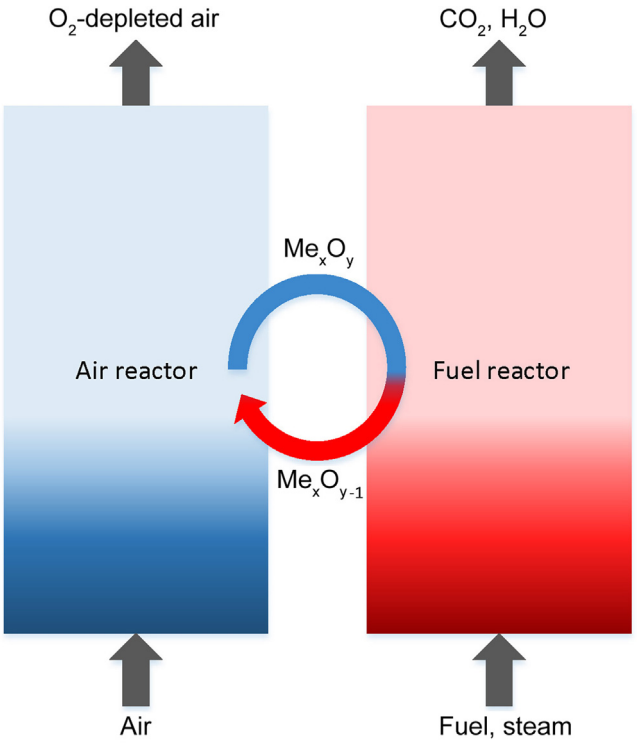

Fig. 1. Schematic drawing of the chemical looping combustion process. The oxygen carrier circulates between the air and fuel reactors, creating a loop. Air is added to one reactor, creating an oxidizing environment, while fuel is added to the other so as to reduce the oxygen carrier.

the effects of changes in the design variables. Such changes may include variations in the geometry of the unit, the operating parameters, or the properties of the solids and gas, within a narrow operational range given by the source measurement data from which they are derived. An example of the correlation-based modeling of CLC is the work of Markström, Berguerand, and Lyngfelt (2010). However, this modeling approach is far from a general method and in fact is highly specific to a particular unit and fuel. Opposite to the empirical correlation-based models are CFD models that are based on fundamental expressions of fluid flow and fluid–particle interactions. These models are general in their formulation and therefore adapt to new geometries, solids, and operational conditions. Thus they are potentially suitable for predictive purposes, such as in the case of new operating conditions or geometries (Adamczyk et al., 2014). Unfortunately, these models are also highly computationally intensive and

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