



Extension and evaluation of a macroscopic model for syngas-fueled chemical looping combustion

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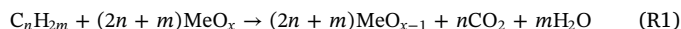
ABSTRACT

Syngas-fueled Chemical Looping Combustion (syngas-CLC) which can be integrated with ex-situ gasification of coal has advantages over the direct use of coal in CLC: (i) no requirement of carbon stripper, (ii) no interaction of oxygen carrier with coal ash, (iii) no loss of oxygen carrier with the draining stream of ash. Few works on simulation of syngas-CLC were performed, although experimental investigations were extensively carried out. In this work, a macroscopic fuel reactor model is extended to a lab-scale 500 W_{th} reactor. The model based on fluid dynamics, mass balance and reduction kinetics is solved with MATLAB® codes and validated against experiments. Influences of various operation parameters are evaluated to study the flexibility of this model. It is shown that the model can give satisfactory predictions for fuel reactor of a syngas-CLC system, independent on the operation conditions. Variations of syngas composition, temperature, solids circulation and oxygen carrier inventory show different effects on flue gas composition and combustion efficiency. After thorough simulation, a region for a combustion efficiency of $\eta_c = 99.9\%$ is proposed, with which the optimized conditions for the 500 W_{th} reactor are established. An oxygen carrier inventory as low as 50 kg/MW_{th} can assure the complete syngas combustion.

1. Introduction

The dependence on fossil fuel for energy production cannot be substituted by other alternatives including renewable energies before they become mature to supply most of the demanded energy [1]. CO₂ emission during the utilization of fossil fuels is considered as one of the major contributors to global warming, thus different strategies for CO₂ removal are developing [2]. Among them, Chemical Looping Combustion (CLC) has been suggested among the most promising technologies for low-cost CO₂ capture [3]. CLC concept is based on the rationale of pure CO₂ generation patented by Lewis and Gilliland [4], which was first proposed by Ishida et al. [5]. In CLC, the conventional combustion is split into two steps: oxygen required for fuel combustion is provided by a solid oxygen carrier, a type of metal oxide; and then air is used to regenerate the oxygen carrier. The most common configuration for CLC is realized by circulating oxygen carrier particles inside interconnected fluidized bed reactors [6]. In the fuel reactor, fuel (C_nH_{2m}) combusts with lattice oxygen of oxygen carrier (MeO_x), while the oxygen carrier

is reduced to MeO_{x-1} as shown in reaction R1. In the air reactor, the previously reduced oxygen carrier MeO_{x-1} is oxidized back to its original form MeO_x by reacting with gaseous oxygen of air via Reaction (R2). It can be seen that CO₂ can be easily separated in Reaction (R1) after a simple condensation of H₂O steam. In this sense, CLC technology provides a prospective technical pathway to address the increase of global CO₂ emission.



For the application to coal combustion, CLC technology was extensively investigated during the past decades [7,8]. Due to the extremely low rate of solid-solid reaction between coal and oxygen carrier particles in fluidization condition [9], several technologies including syngas-fueled Chemical Looping Combustion (syngas-CLC) [10,11], in-situ Gasification Chemical Looping Combustion (iG-CLC) [12] and Chemical Looping with Oxygen Uncoupling (CLOU) [13] were proposed. Syngas-CLC has several advantages over iG-CLC and CLOU,

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Nomenclatures

a	Decay factor of solids concentration in the freeboard
Ar	Archimedes number
b_i	Stoichiometric factor for the reaction of CuO and component i ($i = \text{CO}$ or H_2) in the syngas, mol CuO per mol of gas
C_{ex}	Concentration of gas component at the active surface of oxygen carrier particle, mol m^{-3}
C_i	Concentration of fuel component i ($i = \text{CO}$ or H_2), mol m^{-3}
$C_{j,i}$	Concentration of component i ($i = \text{CO}$, CO_2 , H_2 or H_2O) in bubble ($j = b$), emulsion ($j = e$) or freeboard ($j = f$), mol m^{-3}
C_s	Solids concentration, kg m^{-3}
$C_{s,b}$	Solids concentration at the upper limit of the dense bed, kg m^{-3}
C_z	Bulk gas concentration at height z of the fuel reactor, mol m^{-3}
d_j	Diameter of bubble ($j = b$) or oxygen carrier particle ($j = p$), m
$D_{g,i}$	Diffusivity of gas component i ($i = \text{CO}$, CO_2 , H_2 or H_2O), $\text{m}^2 \text{s}^{-1}$
D_r	Inner diameter of the fuel reactor, m
E_i	Activation energy for the reduction of oxygen carrier by component i ($i = \text{CO}$ or H_2) in the syngas, J mol^{-1}
F_{exc}	Excessive gas molar flow over minimum fluidization, mol s^{-1}
F_j	Gas molar flow at the inlet ($j = \text{in}$) or height z ($j = z$) of fuel reactor or caused by the WGS reaction ($j = \text{WGS}$), mol s^{-1}
$F_{j,i}$	Molar flow of component i ($i = \text{CO}$, CO_2 , H_2 or H_2O) in bubble ($j = b$), emulsion ($j = e$) or freeboard ($j = f$), mol s^{-1}
g	Acceleration of gravity, 9.8 m s^{-2}
H_j	Height of dense bed ($j = b$) or the entire fuel reactor ($j = r$), m
k_{be}	Coefficient for gas exchange between bubble and emulsion, s^{-1}
$k_{g,i}$	Mass transfer coefficient of component i ($i = \text{CO}$ or H_2) in the gas film surrounding oxygen carrier particles, m s^{-1}
k_i	Rate constant of the reaction between component i ($i = \text{CO}$ or H_2) and oxygen carrier, $\text{mol}^{1-n} \text{m}^{3n-2} \text{s}^{-1}$
$k_{o,i}$	Pre-exponential factor of the rate constant for the reaction between component i ($i = \text{CO}$ or H_2) and oxygen carrier, $\text{mol}^{1-n} \text{m}^{3n-2} \text{s}^{-1}$
$K_{\text{WGS,eq}}$	Equilibrium constant for WGS reaction
m	Instantaneous mass of oxygen carrier, kg
m_{FR}	Solids inventory per MW_{th} fuel, $\text{kg MW}_{\text{th}}^{-1}$
m_j	Mass of fully oxidized ($j = \text{ox}$) or reduced ($j = \text{red}$) oxygen carrier, or the bed mass of oxygen carrier in the fuel reactor ($j = s$), kg
M_{O}	Molar weight of oxygen carrier, kg mol^{-1}
\dot{m}_s	Solids circulation rate, kg h^{-1}
n	Reaction order
N_{nz}	Number of nozzle in the gas distributor of fuel reactor
p	Pressure at the outlet of fuel reactor, Pa
P_{th}	Thermal power, W_{th}
Re_p	Reynolds number
R_g	Universal gas constant, $8.314 \text{ J mol}^{-1} \text{ K}^{-1}$

$r_{g,\text{CuO}}$	Radius of CuO grain in the oxygen carrier, m
$(-\bar{r}_{g,i})$	Average reaction rate of gas component i ($i = \text{CO}$ or H_2), $\text{mol m}^{-3} \text{ s}^{-1}$
$(-\bar{r}_{g,i})_j$	Average reaction rate of gas component i ($i = \text{CO}$ or H_2) in emulsion ($j = e$) or freeboard ($j = f$), $\text{mol m}^{-3} \text{ s}^{-1}$
R_{OC}	Oxygen transport capacity
$(-\bar{r}_{s,i})$	Average reaction rate of oxygen carrier particles during the oxidation of component i ($i = \text{CO}$ or H_2) in the syngas, $\text{mol m}^{-3} \text{ s}^{-1}$
Sc	Schmidt number
t	Reaction time, s
T	Temperature, K
t_{mF}	Mean residence time of oxygen carrier particles in fuel reactor, s
u_j	Velocity corresponding to total gas flow ($j = 0$), minimum fluidization flow ($j = \text{mf}$), throughflow ($j = \text{tf}$) or visible bubble flow ($j = \text{vis}$), m s^{-1}
V	Fuel reactor volume, m^3
$V_{\text{M,CuO}}$	Molar volume of CuO grain in the oxygen carrier, $\text{m}^3 \text{ mol}^{-1}$
$(x_i)_j$	Molar fraction of component i ($i = \text{CO}$, CO_2 , H_2 or H_2O) in the gas stream at the inlet ($j = \text{in}$) or height z ($j = z$) of the fuel reactor
$X_{\text{o,in}}$	Oxidation conversion of oxygen carrier at the inlet of fuel reactor
X_s	Solids conversion during reduction
$y_{e,i}$	Molar fraction of component i ($i = \text{CO}$, CO_2 , H_2 or H_2O) in the excessive gas over minimum fluidization
z	Axial position in the fuel reactor, m

Greek Symbols

Δp	Pressure drop over fuel reactor, Pa
δ_b	Bubble fraction in the dense bed
ε_{mf}	Bed porosity at minimum fluidization
ε_s	Solids fraction in fuel reactor
ε_z	Bed porosity at position z of the fuel reactor
η_{C}	Combustion efficiency
ξ_{g-s}	Contact efficiency between gas and solids in freeboard
$\rho_{\text{m,p}}$	Molar density of CuO in the oxygen carrier particles, $\rho_{\text{m,p}} = y_{\text{CuO}}/V_{\text{MCuO}}$, being y_{CuO} the volume fraction of CuO in the oxygen carrier, mol m^{-3}
ρ_p	Particle density, kg m^{-3}
τ_i	Time required for complete reduction of oxygen carrier by gas i ($i = \text{CO}$, H_2 or syn for syngas), s
ψ	Ratio of u_{vis} and $(u_{\text{vis}} + u_{\text{tf}})$
ϕ	Oxygen carrier-to-fuel ratio

Acronyms

CFD	Computational fluid dynamics
CLC	Chemical looping combustion
IGCC	Integrated gasification combined cycle
syngas-CLC	Syngas-fueled chemical looping combustion
SCM	Shrinking core model
TGA	Thermogravimetric analyzer
WGS	Water-gas shift
XRD	X-ray diffraction

namely: no requirement for the carbon stripper, no interaction of oxygen carrier with coal ash and no loss of oxygen carrier with the draining stream of ash. In this sense, syngas-CLC is simpler and more straightforward to be implemented for energy generation from coal.

Syngas-CLC can be easily integrated with Integrated Gasification Combined Cycle (IGCC) process [10,14]. According to the simulations of Jin et al. [10] and Wolf et al. [15], system efficiency of IGCC process can be improved by 5–10% if the conventional CO_2 capture technology

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