

Numerical investigation of chemical looping gasification process using solid fuels for syngas production



Shuai Wang*, Weijie Yin, Zhenjie Li, Xuesong Yang, Kai Zhang

School of Energy Science and Engineering, Harbin Institute of Technology, Harbin 150001, China

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ABSTRACT

Chemical-looping gasification (CLG) is a promising way for syngas production as the integration of chemical looping and gasification technology, which can avoid air separation with a high hydrogen extent in syngas. In this work, the multi-fluid model with chemical kinetics is employed to simulate the CLG performance in a bubbling fluidized bed reactor using char as solid fuels. The distributions of gas compositions and temperatures are predicted. The interactive mechanic between oxygen carrier and solid fuels is analyzed. In addition, the impacts of operating parameters on mixing behaviors of particles and gas products during the CLG process are evaluated. The results demonstrate that the decrease of operating velocity and oxygen carrier particle size will reduce the syngas production. An increase of oxygen carrier size weakens the mixing degree of the binary mixture, whereas increasing the gas inlet velocity can restrict the segregation of the binary mixture.

1. Introduction

Chemical Looping Combustion (CLC) has become a promising novel combustion technology owing to its inherent CO₂ separation and low energy penalty, which uses oxygen carriers to transfer oxygen between air reactor (AR) and fuel reactor (FR) [1,2]. Compared to gaseous fuel, solid fuels like biomass and coal have their potential in abundance and low cost, which have been successfully applied in the CLC system in the last decades [3,4]. In the CLC process with solid fuels, understanding interaction mechanics between fuel and oxygen carriers play an essential role in the promotion of the fuel conversion [5].

Recently, the CLC technology with solid fuels has been extended to produce syngas and hydrogen via integrating the gasification method. As a result of the increase of the H₂/CO ratio, Chemical Looping Gasification (CLG) has attracted more and more concerns [6,7]. Zeng et al. [8] experimentally investigated the CLG performance of sawdust in a dual fluidized bed gasifier and evaluated the impacts of operational parameters. It was pointed out that the dependence of the H₂/CO ratio in the syngas on the FR temperature was significant. Liu et al. [9] examined the effect of the gasifying medium on the CLG process with coal as fuel and CaSO₄ as oxygen carriers. It was found that the reaction mechanism was reverse for the gasifying medium steam and CO₂ although both of them could enhance the syngas production. Huang et al. [10] carried out thermo-gravimetric tests and evaluated the reactivity of different oxygen sources during the CLG process. It was concluded

that NiO has a higher reactivity than steam for char gasification. Meanwhile, it was emphasized that Fe₃O₄ was the main product for reduction reactions of Fe₂O₃.

Computational fluid dynamics (CFD) has been widely applied to investigate the CLC system, which can give some detailed information on how operating parameters influence on the performance of the system [11–14]. Alobaid et al. [15] and Banerjee et al. [16] employed the Eulerian-Eulerian model and Discrete Element Method (DEM) to evaluate the performance of chemical looping combustion process with coal respectively. It was found that the density of oxygen carriers was an important parameter for the enhancement of the CLC performance. Mahalatkar et al. [17] assumed the impacts of high temperature and steam concentration on coal reaction rates during the CLC process based on Eulerian multiphase model. It was demonstrated that the increase of temperature and steam concentration could promote coal reaction rate. Su et al. [18] implemented a simulation of chemical looping combustion with coal in a dual circulation fluidized bed system. The simulated results obtained a reasonable prediction on the measured data. The results revealed that the gas leakage between reactors caused the incomplete char gasification. The flow pattern was dependent on the coal feeding rate. Parker et al. [19] conducted a simulation of the CLC system on the platform of Barracuda VR software and analyzed the efficiency of oxidation and reduction in the air reactor and the fuel reactor. It was shown that there was a poor coal reduction efficiency in the fuel reactor. Up to now, there have been few reports on the

* Corresponding author.

E-mail address: shuaiwang@hit.edu.cn (S. Wang).

Nomenclature

C_d	drag coefficient of a single particle
d	particle diameter [m]
e	restitution coefficient
E	activation energy [KJ mol ⁻¹]
g_0	radial distribution function
k_0	pre-exponential factor
K_i	adsorption constant for component i [Pa ⁻¹]
n	reaction order
P	pressure [Pa]
Re	Reynolds number
R_0	carrying capacity
r_0	grain radius [m]
S_0	initial surface area [m ² m ⁻³]

u	velocity [m s ⁻¹]
X	conversion degree

Greek letters

β	drag coefficient [kg m ⁻³ s ⁻¹]
ε	volume fraction
ρ	density [kg m ⁻³]
ρ_m	molar density of reacting material. [mol m ⁻³]

Subscripts

g	gas phase
s	solid phase

simulation of the CLG process, which requires a further evaluation to favor the hydrogen production.

In the multi-component system like chemical looping system with solid fuels, the discrepancy of physical properties between different solid components results in the mixing and segregation behaviors of particles in the system, which will have a significant influence on the reaction performance of bed materials. In this work, the multi-fluid model is integrated with chemical kinetic models including char gasification and oxygen carrier reduction. A three-dimensional simulation is carried out to investigate the CLG performance with coal as solid fuels and Fe₂O₃ as oxygen carriers in a fluidized bed reactor. The model is validated by the experimental result. The impacts of mixing and segregation behaviors of bi-disperse particles on gasification are analyzed. The effects of operating parameters on the performances are further evaluated.

2. Mathematical model**2.1. Governing equations**

The multi-fluid model is used to describe fluid phase and solid phases in a binary mixture system. Here, it is assumed that each solid phase is spherical with the uniform size and density. The governing equations of the multi-fluid model are summarized in Wang et al. [20], where mass and momentum sources between phases as a result of chemical reactions are taken into account. In order to characterize the particle fluctuating energy, the granular temperature transportation equation is introduced. The kinetic theory of polydisperse particles is employed to close the model [21].

At a high solid volume fraction, the frictional contribution to solid stress should be further considered besides kinetic and collisional contributions. A friction stress model considering the effect of the strain rate fluctuation was developed by Srivastava et al. [22], which is adopted in this work.

2.2. Drag model

Following the investigation of Leboireiro et al. [23], the ad hoc drag treatment can give a similar result as the LBM-based treatment. Hence, the Gidaspow drag model based on the ad hoc drag treatment is adopted in this work [24]. The drag coefficient is expressed as follows:

$$\beta = 150 \frac{(1-\varepsilon_g)^2 \mu_g}{(\varepsilon_g d_s)^2} + 1.75 \frac{\rho_g (1-\varepsilon_g) |u_g - u_s|}{\varepsilon_g d_s} \quad \varepsilon_g \leq 0.8 \quad (1)$$

$$\beta = \frac{3}{4} C_d \frac{\rho_g (1-\varepsilon_g) |u_g - u_s|}{d_s} \varepsilon_g^{-2.65} \quad \varepsilon_g > 0.8 \quad (2)$$

$$C_d = \begin{cases} \frac{24}{Re} (1 + 0.15 Re^{0.687}) & Re \leq 1000 \\ 0.44 & Re \geq 1000 \end{cases} \quad (3)$$

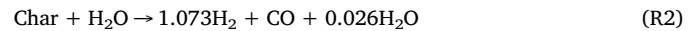
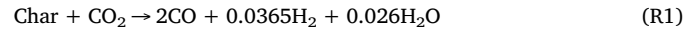
Here, some modifications are made in the ad hoc drag treatment. The particle diameter and the slip velocity are replaced by the species diameter and the species slip velocity. It is assumed that the drag law depends on the total solid volume fraction rather than the composition of the polydisperse mixture [23].

In order to characterize the interaction between solid phases, the solid–solid drag coefficient proposed by Syamlal-O'Brien is applied [25]:

$$\beta_{lm} = \frac{3(1 + e_{lm})(\pi/2 + C_{lm}\pi^2/2)(d_l + d_m)^2 \varepsilon_l \rho_l \varepsilon_m \rho_m g_{0,lm} |u_l - u_m|}{2\pi(\rho_m d_m^3 + \rho_l d_l^3)} \quad (4)$$

2.3. Chemical reaction kinetic model

Chen et al. [26] conducted an experimental investigation of chemical-looping gasification in a bench-scale fluidized bed facility, where the synthetic Fe₂O₃/Al₂O₃ was chosen as oxygen carriers and the coal char was used as solid fuel, which is adopted as the objective of this work. The proximate and ultimate analyses of coal char can be found in Table 1 [26]. The main reactions are given as follows:

1. Char gasification:

Here, the char gasification rate takes the kinetic model of Everson et al. [27] and is expressed as below:

$$\dot{m}_{\text{Char}} = \rho_s \varepsilon_s \frac{S_0}{1-\varepsilon_0} r (1-X_s)^{2/3} \quad (5)$$

where S_0 represents char initial surface area. For the two gasification agents, the gasification rates are written as:

Table 1
Ultimate and proximate analysis of coal char [26].

Ultimate analysis	wt%	Proximate analysis	wt%
C	76.7	Ash	14.72
H	1.84	Volatiles	4.34
O	2.55	Fixed carbon	80.94
N	1.65		
S	2.48		

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