Full Length Article

In situ experimental and modeling study on coal char combustion for coarse particle with effect of gasification in air (O₂/N₂) and O₂/CO₂ atmospheres

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A B S T R A C T

This study applied the high temperature stage microscope to investigate coal char combustion in air (O₂/N₂) and O₂/CO₂ (volume, 21%/79%) atmospheres with the effect of gasification. A reaction front combustion model was also proposed, coupled with effect of char gasification, to predict the key parameters of char combustion in the O₂/CO₂ atmospheres. The combustion process and particle evolution of coarse chars were observed and measured to calculate the carbon conversion. Experimental results showed that the burnout time of the char particle in the O₂/CO₂ atmosphere in the was prolonged about 20–25%, compared to the burnout time of chars in air. The overall reaction rates of char particles in the O₂/CO₂ atmosphere were lower than the rates in the air for different particle sizes. In addition, reaction rates from model prediction showed good agreements with the experimental data for different particle sizes both in the air and O₂/CO₂ atmospheres, which proved the reaction front combustion model was applicable for the char combustion in this study. The reaction front combustion model also predicted that the reaction front decreased with burning time and mass consumption of char particle. For the char combustion in the O₂/CO₂ atmosphere, the gasification with CO₂ absorbed part of the combustion heat or radiation heat and caused lower particle temperatures and heat fluxes for different particle sizes. The proposed combustion model also predicted that although the effect of the gasification reaction was dominant at the beginning of combustion and then reduced, the conversion of particles was hindered compared to the combustion of char in air.

1. Introduction

Oxy-fuel combustion technology in the coal-fired furnaces is one of the promising applications for CO₂ capture, lower emission of NOx and other pollutants [1,2]. This technology involves the combustion of pulverized coal in the mixture of oxygen and recirculated flue gas (mainly CO₂) in order to reduce the net volume of flue gas from the process and to substantially increase the concentration of carbon dioxide (CO₂) in the flue gas, and the CO₂ recirculation is to moderate the temperature in the combustion and make up the volume of the missing N₂ to ensure there is enough gas to carry the heat through the boiler during oxy-fuel combustion [3]. Coal combustion in O₂/CO₂ atmosphere was different from the combustion in air, such as particle ignition, flame character heat transfer behavior, combustion characteristics and generated pollutants, with effects of coal rank, single particle

character, oxygen concentration and pressure, etc. [4–7]. Differences between oxy-fuel combustion and traditional combustion were mainly due to the replacement of N₂ to CO₂ with physical and chemical effects on the combustion.

Compared to the properties of N₂ in air atmosphere, CO₂ with larger density and molar heat capacity in the mixture gases of O₂ and CO₂ would cause the thermophysical effects on the coal/char combustion during oxy-fuel combustion. Liu et al. [8] found that simply replacing N₂ with CO₂ resulted in the decreased temperature of combustion gas and the delayed char burnout time, owing to the larger specific heat capacity of CO₂. In the O₂/CO₂ atmosphere, the ignition of coal particle occurred as a form of volatile cloud rather than individual particles that occurred in air, and the temperature of volatile flame and char particles were also reduced by CO₂ quenching throughout coal oxidation [9]. Levendis et al. [4,10] similarly pointed out that the reaction gas

Nomenclature

A_t	cross-sectional area at time t (m^2)
A_0	initial cross-sectional area (m^2)
A_p	particle surface area (m^2)
A_I	pre-exponential factor for heterogeneous oxidation ($kg\,m^{-2}\,s^{-1}\,Pa^{-n}$)
A_{II}	pre-exponential factor for heterogeneous gasification ($kg\,m^{-2}\,s^{-1}\,Pa^{-n}$)
D	diffusion coefficient ($m^2\,s^{-1}$)
E_I	activation energy for heterogeneous oxidation ($J\,mol^{-1}$)
E_{II}	activation energy for heterogeneous gasification ($J\,mol^{-1}$)
f_g	force from gas flow (N)
f_s	friction force (N)
G	mass flux ($kg\,m^{-2}\,s^{-1}$)
H_I	heat of heterogeneous oxidation $C + 1/2O_2 \rightarrow CO$ ($J\,kg^{-1}$)
H_{II}	heat of heterogeneous gasification $C + CO_2 \rightarrow 2CO$ ($J\,kg^{-1}$)
H_{III}	heat of heterogeneous oxidation $C + O_2 \rightarrow CO_2$ ($J\,kg^{-1}$)
K	mass transfer coefficient ($kg\,m^{-2}\,s^{-1}$)
m_t	particle mass at time t (kg)
m_0	initial particle mass (kg)
m_p	particle mass (kg)
M	molar mass ($g\,mol^{-1}$)
n	reaction order
P	pressure (Pa)
q_C	overall reaction rate of particle surface ($kg\,m^{-2}\,s^{-1}$)
q_O	surface reaction rate for heterogeneous oxidation ($kg\,m^{-2}\,s^{-1}$)
q_P	surface reaction rate for heterogeneous gasification ($kg\,m^{-2}\,s^{-1}$)
Q	heat flux ($W\,m^{-2}$)
Q_r	radiation heat flux ($W\,m^{-2}$)
r	particle radius (m)
r_s	radius at particle surface (m)
r_f	radius at reaction front (m)
R'	universal gas constant ($m^3\,Pa\,mol^{-1}\,K^{-1}$)
t	time (s)
T	temperature (K)
T_g	gas temperature (K)
T_s	temperature at particle surface (K)
T_f	temperature at reaction front (K)
T_w	wall temperature (K)
u	particle velocity ($m\,s^{-1}$)
x	carbon conversion

Greek symbols

ρ	density ($kg\,m^{-3}$)
δ	thickness of reaction front (m)
ε	emissivity
λ	thermal conductivity ($W\,m^{-1}\,K^{-1}$)
σ	Stefan-Boltzmann constant ($W\,m^{-2}\,K^{-4}$)
φ	molar mass ratio

Subscripts

0	at initial time t = 0
d	diffusion
f	reaction front
F	carbon monoxide (CO)
g	gas
O	oxygen (O_2)
p	char particle
P	carbon dioxide (CO_2)
s	particle surface
t	time
w	wall of furnace
I	heterogeneous oxidation $C + 1/2O_2 \rightarrow CO$
II	heterogeneous gasification $C + CO_2 \rightarrow 2CO$
III	heterogeneous oxidation $C + O_2 \rightarrow CO_2$
∞	ambience

changing from air to the oxy-fuel atmosphere at analogous oxygen mole fractions reduced the combustion temperatures and lengthened the burnout times of coal particles. In addition, researchers also found that oxygen with lower binary molecular diffusion in CO_2 reduced the O_2 flux to the surface and internal of particle resulting in the decrease of reactivity [10,11]. Molina and Shaddix [12] proposed a detailed one-dimensional transient model with comparison of measurements in the air and O_2/CO_2 atmospheres and predicted the similar combustion parameters (e.g. delayed ignition time, lower flame temperature and char temperature) in the higher condition of CO_2 with a lower O_2 concentration.

However, researchers also found the high concentration of CO_2 around coal/char particle in oxy-fuel combustion enhanced the oxidation of char via the gasification reaction, which decreased overall surface reaction rate of char combustion [13–15]. Due to the insufficient O_2, reactions for partial oxidation and even char gasification to CO were occurred in char particle vicinity [3,9]. Then a thin layer of CO product around coal/char particle reduced the burning rate due to consumption of oxygen, allowing for less of O_2 to reach the particle surface, which became influential at higher temperature and pressure with high concentrations of CO_2. [15]. An investigation by Hecht et al. [16] for a single particle combustion in the $O_2/CO_2/H_2O$ mixtures showed that the gasification reaction accounted for roughly 20% of the carbon consumption in the combustion of low oxygen condition, and for about 30% under oxygen-enriched condition. Zhou et al. [17] also found that carbon conversion region at high temperature exhibited large gasification rate while low-temperature region showed weak gasification reaction.

To study the combustion characteristics and predict the key parameters of coal/char particles in the oxy-fuel combustion, models depending on the different assumptions of reaction mechanism were also proposed with the thermophysical and chemical effects on combustion, which were generally classified into single-film [18,19], double-film [20,21] and continuous-film models [18,22–24]. Among these models, the continuous-film model pointed out that homogeneous CO oxidation happened on char particle surface at a low concentration of oxygen in the stagnation flow [25,26]. Zhang et al. [27] proposed a combustion model with a moving flame front and considered the effect of CO oxidizing in the boundary layer of particle combustion. Makino and Law [28] conducted a fairly comprehensive modeling investigation and found that when the temperature for sufficiently large particle continuously increased in O_2/CO_2 atmosphere, gasification actually involved an initial particle heating period. Hecht et al. [16,21,29] and Kim et al. [30] also modeled the char combustion and found gasification had increasing influence on char combustion with the increasing temperature and particle size. Besides, notable effects of Stefan flow and CO oxidation on the particle burnout time under lower O_2 concentration in oxy-fuel combustion exhibited with lower reactivity, lower gas temperature and larger particle size [31,32]. The particle size of grinding coal powder was not homogeneous, there was still a part of large particles (more than 100 μm) existed in the pulverized coal. Larger particles additionally have a greater capacity for storing energy