Numerical investigation on separate physicochemical effects of carbon dioxide on coal char combustion in O_2/CO_2 environments

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ABSTRACT

The thermophysical and chemical effects of CO_2 on the combustion characteristics of a single char particle in O_2/CO_2 environments remain unclear because these effects are intercoupled during the complex process of coal char combustion. A numerical method was proposed to adjust the char particle temperature in an O_2/CO_2 environment back to that in an O_2/N_2 environment by replacing an appropriate portion of CO_2 with argon because of the molar heat capacity of Ar $< N_2 < CO_2$, and to quantitatively isolate the effects of CO_2 on bituminous coal char combustion in specially designed combustion environments with the continuous-film model. The numerical result shows that the char particle temperature in a 21% O_2/79% CO_2 environment is lower by 493 K than that in a 21% O_2/79% N_2 environment because of the combined effects of lower local gas temperature and endothermic char gasification reaction with CO_2. The char particle temperature increases significantly with the oxygen concentration, and the char temperature in a 52.5% O_2/47.5% CO_2 environment is equivalent to that in air environment. Most importantly, the effects of CO_2 on bituminous coal char combustion in O_2/CO_2 environments are quantitatively isolated with specially designed combustion environments (21% O_2/79% N_2, 21% O_2/11% CO_2/68% Ar, 52.5% O_2/47.5% CO_2 and 52.5% O_2/30% CO_2/17.5% Ar). The relative contributions of the oxygen concentration, thermal, and chemical effects on the char combustion rate are 82.1%, 11.2%, and 6.7%, respectively, at an ambient gas temperature of 1200 K for bituminous coal char of 91 μm. The results show that the oxygen concentration effect is the most important factor, followed by the thermal effect and the chemical effect of char gasification reaction with CO_2.

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1. Introduction

Oxy-fuel combustion is regarded as one of the most promising technologies for CO_2 capture [1,2]. Oxy-coal combustion is greatly altered in coal ignition, flame stabilization and char combustion because of the different physicochemical properties of CO_2 compared with conventional pulverized coal combustion in air environment. The density and molar heat capacity of CO_2 are much higher than those of N_2, and the diffusivity of O_2 in the O_2/CO_2 mixture is lower than that in the O_2/N_2 mixture. In addition, CO_2 is the reactant of char consumption through the char gasification reaction with CO_2, which is distinct from N_2 as an inert oxidant [3–6]. These thermophysical and chemical effects of CO_2 on the combustion characteristics of a single char particle in O_2/CO_2 environments remain unclear because these effects are intercoupled during the complex process of coal char combustion and it is difficult to elucidate these separate effects of CO_2 on char combustion.

A number of oxy-coal combustion investigations have focused on the overall CO_2 effect on the coal ignition delay time, char particle temperature and char combustion rate. Molina and Shaddix [7,8] used a laminar entrained flow reactor to investigate coal ignition and devolatilization time with an intensified CCD camera in different O_2/CO_2 and O_2/N_2 environments. Their results showed that coal ignition was delayed in O_2/CO_2 environments with high concentrations of CO_2 compared with that in air environment. Levendis and co-workers [9–11] monitored the time-dependent combustion histories of a single coal particle in different environments with a calibrated three-color pyrometry and revealed that the coal particle temperature was lower in an O_2/CO_2 environment than in an O_2/N_2 environment at the same oxygen concentration and that the corresponding char burnout time was longer. Zhang et al. [12,13] demonstrated that the coal char particle temperature decreases because of CO_2 quenching during the char combustion process. They also observed that the char gasification reaction with CO_2 improved the coal mass reduction rate. Hence, we can infer from these previous results that the combined effects of different particle temperatures and oxygen concentrations

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Nomenclature

C_P	specific heat (J/(kg·K))
D	diffusion coefficient (m²/s)
Da	Damköhler number
E	activation energy (J/mol)
H	heat of reaction (J/kg)
k	reaction rate coefficient (m/s)
m	combustion rate (kg/s)
M	molecular weight (g/mol)
Q	stoichiometric carbon-to-CO reaction heat ratio
r	radial distance from carbon particle (m)
R_0	universal gas constant (J/(mol·K))
R	transient-to-initial radius ratio
t	time (s)
T	temperature (K)
w	reaction rate (kg/(m²·s) or (kg/m³·s))
Y	mass fraction
ρ	density (kg/m³)
υ	stoichiometric coefficient
ε	emissivity of particle surface
τ	non-dimensional time
θ	non-dimensional activation energy
σ	Stefan–Boltzmann constant (W/(m²·K⁴))
λ	Thermal conductivity, (W/(m·K))
α	stoichiometric CO_2-to-reactant mass ratio
β	conventional transfer number
γ	temperature gradient at the surface
δ	product(CO_2)-to-carbon mass ratio
ξ	profile function

Subscripts

Ar	Ar
C	carbon
F	CO
g	gas
N	N_2
O	O_2
P	CO_2
s	particle surface
∞	ambience
0	initial state
1	reaction C+1/2O_2→CO
2	reaction C+CO_2→2CO

Superscript

~	dimensionless quantity

cant factor that caused flame liftoff. Recently, Al-Qurashi et al. [17] successfully isolated the thermal, dilution, and chemical effects of CO_2 on soot reactivity in the exhaust gas recirculation of a diesel engine and concluded that the thermal effect of CO_2 was the most important factor in soot formation.

Similarly, the different effects of CO_2, including the oxygen concentration, thermal, and chemical effects of char gasification reaction with CO_2, on oxy-coal combustion are intimately coupled. It is difficult to experimentally separate the changes in char particle temperature and combustion rate caused by the different thermophysical and chemical effects of CO_2. However, numerical simulation is a feasible method to separate these effects of CO_2 on char combustion. Depending on different assumptions of reaction mechanism for both particle surface and gas phase chemistry, char combustion models are generally classified into single-film, double-film and continuous-film models. The continuous-film model [18,19] simultaneously accounts for the surface oxidation and reduction and the CO oxidation reaction in the boundary layer of char particle and faithfully reflects the char combustion process. Makino et al. [20,21] derived the coupling functions for carbon particle combustion and simulated the quasi-steady and transient combustion behavior of a carbon particle in an O_2/CO_2/inert environment. Hecht et al. [22–24] and Kim et al. [25] investigated the effect of CO_2 and steam gasification reactions on coal char combustion using the kinetics code SKIPPY and the extended single-film model and revealed that CO_2 gasification reactions increased the char combustion conversion rate at low oxygen concentration and decreased the char combustion conversion rate at high oxygen concentration [22]. Yu et al. [26] investigated the effect of Stefan flow on char particle combustion in an O_2/CO_2 atmosphere and concluded its notable effect on burnout time at lower oxygen concentration. Nevertheless, they both considered the continuous-film model to be a more accurate benchmark for predicting the oxy-coal combustion process [25–27].

The objective of the current study is to present a numerical method to adjust the char particle temperature in O_2/CO_2 environments back to that in O_2/N_2 environments without changing the oxygen concentration by replacing an appropriate portion of CO_2 with Ar because argon has a lower molar heat capacity (e.g. 21 kJ/(kmol·K) at a gas temperature of 1200 K) than nitrogen (33 kJ/(kmol·K)) and carbon dioxide (56 kJ/(kmol·K)) at the same gas temperature. The different effects of CO_2 on the char temperature and combustion rate in the comparable O_2/CO_2, O_2/ CO_2/Ar, and O_2/N_2 environments are then quantitatively isolated using the continuous-film model, and the numerical results are analyzed to elucidate the separate effects of CO_2 on the underlying mechanism of char combustion in O_2/CO_2 environments.

2. Model

2.1. Model description

The continuous-film model assumes that a char particle is spherical and burns in a quiescent O_2/CO_2/N_2/Ar mixture, as shown in Fig. 1. The primary product of the heterogeneous oxidation on the surface of the char particle is mainly CO when a single char particle burns in a high-temperature ambience (i.e., higher than 1000 K) [20].

Consequently, the dominant char surface reactions are

$$2C + O_2 \rightarrow 2CO \tag{I}$$

$$C + CO_2 \rightarrow 2CO \tag{II}$$

The further oxidation of CO in the boundary layer of char particle is

on the char combustion rate are coupled with the char gasification reaction with CO_2 during the actual oxy-coal combustion process.

Moreover, some researchers have investigated the influence of carbon dioxide additive on soot formation on both the fuel and oxidizer sides of a diffusion flame and isolated the effects of dilution, temperature, and chemical participation to understand the relative influence of these mechanisms [14]. CO_2 has been noted to be capable of triggering the dilution (low oxygen concentration), thermal, and chemical effects on the fuel combustion process when introduced into the fuel or oxidizer side. Du et al. [14] presented a method to adjust the same flame temperature without changing the oxygen concentration by replacing a portion of CO_2 in the oxidizer with argon (Ar) and isolated the dilution, thermal, and chemical effects of CO_2 on soot formation in diffusion flames. Liu et al. [15] and Guo et al. [16] numerically simulated the effects of CO_2/N_2/Ar addition into air in a laminar diffusion flame and revealed that the dilution effect of CO_2 was the most signifi-