Original Research Paper

Numerical simulation of soot formation in pulverized coal combustion with detailed chemical reaction mechanism

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A R T I C L E   I N F O

A B S T R A C T

A two-dimensional unsteady numerical simulation with a detailed chemical reaction mechanism that considers 158 species and 1804 reactions is applied to pulverized coal combustion in a mixing layer and the soot formation behavior is investigated in detail. The computational conditions and ignition process are the same as those in our previous work (Muto et al., 2017). The results show that the peak of the mass density of the soot is distributed in the region where the gas temperature is higher than the unburned gas temperature of the mixture of volatile matter and air (1300–1400 K) and lower than the flame temperature (2000 K $\sim$). This is due to the fact that soot formation from the precursors (C_2H_2 and C_6H_6) is enhanced as the gas temperature increases, whereas the quantities of the precursors and the produced soot are reduced due to oxidation at the higher gas temperature condition that exists close to the flame. The peak value of the mass density of the soot is also distributed in the region between the peak values of the gas temperature and the probability density function of the number of coal particles.

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1. Introduction

Coal is the primary energy source worldwide because of its low procurement costs, which can be attributed to its numerous widespread recoverable reserves and stable supply. However, because coal-fired power generation produces large amounts of pollutants such as soot as well as sulfur oxide or nitrogen oxide, it is important to improve the combustion efficiency and to reduce the burden on the environment if we wish to continue using coal as a major source of energy in the future. The combustion process that occurs within the pulverized coal boiler is extremely complex as it involves several elementary processes such as the dispersion of particles in the flow field, heat transfer between the particles and the air surrounding them, release of flammable gases (volatile matter) from the particles, solid-state reactions, and gas-phase reactions, all of which are greatly affected by turbulence. Because of this complexity, employing numerical simulations is very helpful in designing and developing furnaces efficiently to obtain details that cannot be obtained from experiments.

Recent computational fluid dynamics methods such as large-eddy simulation (LES, e.g., [1–8]) and direct numerical simulation (DNS, e.g., [9–13]) for the carrier gas have been applied to coal combustion and have revealed aspects of combustion behavior such as flamelet and ignition characteristics. So far, a few soot formation models in coal combustion classified into empirical model [14], semi-empirical model [15–17], detailed model [18] have been developed. Among the above models, the semi-empirical model has been further improved in recent years [19] due to the relatively low prediction accuracy of the empirical model and the high calculation cost of the detailed model. However, most of these models are applied to soot formation during the combustion of a single pulverized coal, and there are only a few studies [20] in which above physical models are applied to pulverized coal combustion field such as a coal jet flame to discuss the soot formation process. Although Hayashi et al. [21] recently measured the spatial distribution of the soot volume fraction using laser induced incandescence (LII) in the coal-air jet flame, the obtained data are extremely limited to understand the soot formation mechanism.

The purpose of this study is, therefore, to obtain a physical insight into the chemical reaction in the soot formation process of mono-dispersed pulverized coal particle-laden flow using two-dimensional unsteady numerical simulation with detailed chemical reaction mechanism.

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2. Numerical methods

2.1. Governing equations

The numerical methods for the present two-dimensional unsteady numerical simulation are basically the same as those in Muto et al. [13]. The governing equations for the two-dimensional unsteady numerical simulation of the gas phase are the conservation equations of mass, momentum, energy, and mass fraction of each chemical species, and the equation of state for the ideal gas. While, the equations for the dispersed phase (coal particles) are the equations for each coal particle position, velocity, temperature, and mass. The shape of the coal particle is assumed to be a spherical, and collision and breakup of particles are not considered. It is also assumed that the change in particle mass leads only to change in density while the particle diameter remains constant. Radiation flux in the energy equation is calculated by the discrete ordinate method (DOM)/S4 [22] with a coefficient of absorption obtained by the weighted sum of gray gases (WSGG) [23]. The equations for the soot are the transport equations of the soot number density and the mass density, which are described later.

These equations are solved using an in-house thermal flow analysis code referred to as FK^3 (e.g., [10,12,13,24,25]). The interactions attributed to the phase coupling between the gas and the dispersed-coal phases are calculated by a Particle-Source-In-Cell (PSI-Cell) model [26].

2.2. Coal combustion modeling

Heated coal particles yield char containing fixed carbon, nitrogen,ash, and volatile matter upon thermal decomposition. In the present study, the devolatilization rate and compositions of the volatile matter of coal are obtained using basically the same manner as Hara et al. [10] and Muto et al. [12]. The devolatilization rate of volatile matter is modeled as

$$\frac{dV}{dt} = K_v(V^* - V'), \tag{1}$$

$$K_v = A_v \exp\left(-\frac{E_v}{RT_p}\right). \tag{2}$$

Here, V is the volatile matter mass, and V^* and V' the initial volatile matter content in coal and the volatilized mass released from coal, respectively. Kinematic parameters of frequency factor, A_v, and activation factor, E_v, are separately given by 2.4×10^4 s^{-1} and 3.7×10^4 J/mol, respectively, for each chemical species in volatile matter, and 2.0×10^4 s^{-1} and 3.1×10^4 J/mol, respectively, for

Table 1
Properties of Newlands coal [10].

High heating value	29.1 MJ/kg
Low heating value	28.1 MJ/kg
Proximate analysis (wt.%)	
Moisture[a]	2.60
Ash[b]	15.20
Volatile matter[b]	26.90
Fixed carbon[b]	57.90
Ultimate analysis[b] (wt.%)	
Carbon	71.90
Hydrogen	4.40
Nitrogen	1.50
Oxygen	6.53
Combustible sulfur	0.39

[a] As received.
[b] Dry basis.

Table 2
Composition of volatile matter obtained using NLG version of CPD model [27].

Species	wt%
H_2	0.84
H_2O	10.92
CO	6.37
CO_2	4.06
CH_4	9.68
C_2H_4	2.51
C_2H_6	2.51
C_3H_6	2.51
C_3H_8	1.25
C_6H_6[a]	59.34

[a] C_6H_6 is assumed to represent tar in this study.

H_2O. The chemical reaction model for gas phase for the present two-dimensional unsteady numerical simulation is basically the same as that in Muto et al. [13] In this study, the coal sample is Newlands [10], the properties of which are listed in Table 1 and the compositions of the coal particle, which is obtained using the nitrogen and light gas (NLG) version of the chemical percolation devolatilization (CPD) model [27], are listed in Table 2. It should be noted that in this paper, tar is assumed to be composed of C_6H_6 (benzene). The fixed carbon in the char begins to oxidize at the same time as the commencement of devolatilization. The change in the mass of the char associated with the oxidation reaction is estimated using the Field model [28] by

$$\frac{dC}{dt} = -\left(\frac{K_c K_d}{K_c + K_d}\right) P_{O_2} \pi d_p^2, \tag{3}$$

$$K_c = A_c \exp\left(-\frac{E_c}{RT_p}\right), \tag{4}$$

$$K_d = \frac{5.06 \times 10^{-7}}{d_p}\left(\frac{T_p + T}{2}\right)^{0.75}, \tag{5}$$

where both the chemical reaction rate and the O_2 diffusion rate to solid surface are considered. Here, C is the char mass, P_{O_2} the partial O_2 pressure, d_p the coal particle diameter, K_c and K_d the chemical and diffusion rate coefficients, and T_p and T the coal particle temperature and gas temperature, respectively. The values of the kinematic parameters of the frequency factor, A_c, and activation energy, E_c, are 6.0×10^{-3} s/m and 5.0×10^4 J/mol, respectively [29].

2.3. Soot formation modeling

The transport equations for the soot number density, N, and the mass density, M, are given as

$$\frac{\partial \rho \phi_N}{\partial t} + \nabla \cdot (\rho \phi_N \boldsymbol{u}) = \nabla \cdot (\rho D_{soot} \nabla \phi_N) + \nabla \cdot \left(\gamma \frac{\mu}{T} \phi_N \nabla T\right) + S_N, \tag{6}$$

$$\frac{\partial \rho \phi_M}{\partial t} + \nabla \cdot (\rho \phi_M \boldsymbol{u}) = \nabla \cdot (\rho D_{soot} \nabla \phi_M) + \nabla \cdot \left(\gamma \frac{\mu}{T} \phi_M \nabla T\right) + S_M, \tag{7}$$

where ρ is the density, $\boldsymbol{u}$ the gas velocity, μ the viscosity, ϕ_N the mass molar concentration of soot, ϕ_M the mass fraction of soot, and D_{soot} the diffusion coefficient of soot. Here, N and M are given by

$$N = N_A \rho \phi_N, \quad M = \rho \phi_M. \tag{8}$$

N_A is the Avogadro's number ($N_A = 6.02 \times 10^{23}$ mol^{-1}). The second term on the right-hand side in the expressions for ϕ_N and ϕ_M accounts for the thermophoretic transport, where