Applied Thermal Engineering 63 (2014) 565–576

Contents lists available at ScienceDirect

Applied Thermal Engineering

journal homepage: www.elsevier.com/locate/apthermeng

Intrinsic reaction kinetics of coal char combustion by direct measurement of ignition temperature

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HIGHLIGHTS

- A novel technique was used to measure the coal char particle temperature.
- The ignition point determined from a dT/dt minimum is a spot ignition point.
- A spot ignition model was suggested to analyze the intrinsic reaction kinetics of coal char.
- Internal conduction has to be considered in order to evaluate the intrinsic kinetics for larger particle (above 1 mm).

ARTICLE INFO

Article history:
Received 15 July 2013
Accepted 25 November 2013
Available online 4 December 2013

Keywords:
Coal char combustion
Intrinsic reaction kinetics
Wire heating reactor
External/internal oxygen diffusion
Internal conduction

ABSTRACT

A wire heating reactor that can use a synchronized experimental method was developed to obtain the intrinsic kinetics of large coal char particles ranging in size from 0.4 to 1 mm. This synchronization system consists of three parts: a thermocouple wire for both heating and direct measurement of the particle temperature, a photodetector sensor for determining ignition/burnout points by measuring the intensity of luminous emission from burning particles, and a high-speed camera–long-distance microscope for observing and recording the movement of luminous zone directly. Coal char ignition was found to begin at a spot on the particle's external surface and then moved across the entire particle. Moreover, the ignition point determined according to the minimum of dT/dt is a spot point and not a full growth point. The ignition temperature of the spot point rises as the particle diameter increases. A spot ignition model, which describes the ignition in terms of the internal conduction and external/internal oxygen diffusion, was then developed to evaluate the intrinsic kinetics and predict the ignition temperature of the coal char. Internal conduction was found to be important in large coal char particles because its effect becomes greater than that of oxygen diffusion as the particle diameter increases. In addition, the intrinsic kinetics of coal char obtained from the spot ignition model for two types of coal does not differ significantly from the results of previous investigators.

1. Introduction

Coal, which is clearly an essential energy resource for meeting future energy needs, accounts for about 40% of the electricity generated worldwide. In Korea, it accounted for 38.4% of all energy resources consumed to generate electrical power in 2007 according to data from the Korea Energy Economics Institute [1]. Although clean coal technologies have been introduced, most coal-powered electricity is generated by direct coal combustion technologies such as pulverized fuel fired boiler (PF) and circulating fluidized bed (CFB) power plants [2,3]. A key technology in such plants is temperature control for purposes of heat exchange. Moreover, most power plants in Korea generate power using non-designed coal, which is of low rank, for the boiler because the low rank coal is imported owing to a recent increase in the price of high rank coal. When non-designed coal is used, combustion problems arise owing to its fuel characteristics that make it difficult to obtain the optimal operating conditions. Therefore, the combustion kinetics of coal continues to attract attention because it not only provides basic information on the burnout time and temperature required for optimal combustor design, but also significantly affects the thermal efficiency of combustion in power plants.

Coal combustion has five well-defined steps: heating, moisture evaporation, devolatilization, volatile combustion, and char combustion [4]. Char combustion is particularly important because it determines the thermal efficiency and burnout time in the boiler. Previous investigators have used different experimental methods

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1359-4311/$ – see front matter © 2013 Published by Elsevier Ltd.
http://dx.doi.org/10.1016/j.applthermaleng.2013.11.055

Nomenclature

Capital letters

A	pre-exponential factor (g/cm^2/s)
Bi	Biot number (unitless)
C_{O_2}	oxygen concentration (mol/cm^3)
E	activation energy (kJ/mol)
D_e	molecular diffusion coefficient (cm^2/s)
$D_{eff,p}$	effective diffusion coefficient (cm^2/s)
I	current of the circuit (A)
L	self-inductance ($H = \Omega$ s)
M_c	carbon molecular weight (g/mol)
Nu	Nusselt number (unitless)
$\dot{N}$	molar oxygen diffusion rate (mol/s)
Pr	Prandtl number (unitless)
$P_{O_2,s}$	oxygen partial pressure at the particle surface (Pa)
$\dot{Q}_{cond}$	conduction heat rate (J/s)
$\dot{Q}_{conv}$	convection heat rate(J/s)
$\dot{Q}_G$	generated heat rate(J/s)
$\dot{Q}_L$	heat loss rate(J/s)
$\dot{Q}_{rad}$	radiation heat rate(J/s)
Ra	Rayleigh number (unitless)
R_u	universal gas constant (J/mol/K)
R_Ω	resistance of helical shape thermocouple (Ω)
S_g	specific surface area (cm^2/g)
T_c	center temperature of particle (K)
T_g	ambient gas temperature (K)
T_{ig}	ignition temperature (K)
T_s	particle surface temperature (K)
T_{TC}	thermocouple junction temperature (K)
V	voltage of the circuit (V)

Lowercase letters

d_p	coal char particle diameter (cm)
d_{TC}	helical shape diameter of thermocouple (cm)
g	acceleration of gravity (cm/s^2)
h_c	heat of combustion (J/g)
h	convection coefficient (J/s/K/cm^2)
k	intrinsic rate constant (s cm/g)
k_a	apparent kinetics (1/s/Pa)
k_{char}	conduction coefficient of coal char (J/s/K/cm)
k_i	intrinsic kinetics (1/s/Pa)
k_{cond}	conduction-modified kinetics (1/s/Pa)
m	mass of single char particle (g)
r	radius from the particle center to a point inside a particle layer (cm)
r_p	outer radius of primary particle (cm)
r'_c	char consumption rate (g/s)
x	duty fraction for controlling an amount of initial voltage (unitless)

Greek letters

$\acute{a}$	thermal diffusivity (cm^2/s)
$\hat{a}$	molar stoichiometric coefficient (unitless)
$\ddot{a}$	thermal expansion diffusivity (1/K)
$\mathring{a}$	particle emissivity (unitless)
ς_p	effectiveness factor of inside diffusion (unitless)
ς_e	effectiveness factor of external diffusion (unitless)
$\ddot{e}$	thermal conductivity of gas (J/cm/s/K)
$\acute{\imath}$	kinematic viscosity (cm^2/s)
$\acute{o}$	Stefan–Boltzmann constant (J/cm^2/s/K)
$\tilde{n}_{b,p}$	bulk density of a particle (g/cm^3)
ρ_{TC}	thermocouple density, Pt/PtRh 13% (g/cm^3)
$\hat{o}_p$	tortuosity (unitless)
$\hat{O}_p$	Thiele diffusion modulus (unitless)
$\ddot{o}_p$	porosity (unitless)
ψ	fraction of CO_2 in a reaction product (unitless)

Acronyms

HSC–LDM	high-speed camera–long-distance microscope
PDS	photodetector sensor
WHR	Wire heating reactor

for evaluating the combustion rate of char particles [5–11]. These methods enable the analysis of the combustion rate at ambient temperatures normally ranging between 1200 and 2000 K [12–17]. These experimental methods are classified broadly into energy balance methods based on the particle temperature and size, and mass balance methods based on the particle size, particle mass, and CO/CO_2 concentrations. The particle temperature is especially important because the kinetics are exponentially related to the temperature. Semenov's thermal spontaneous ignition theory [18] has been applied by previous investigators [19–23] to analyze the kinetics according to the energy balance method because the mass of coal char after all volatiles have been emitted is unchanged until the moment it ignites.

The theoretical analysis of char combustion based on the above-mentioned previous investigations is challenging because of changes in the reaction rate regimes depending on the particle temperature and mass transfer during combustion. The coal char combustion rate can be classified as belonging to three regimes: regime III, in which the bulk surface diffusion controls the overall particle reaction rate [24]; regime II, in which the combined effects of the pore diffusion and intrinsic chemical reactivity control the overall particle reaction rate; and regime I, in which the intrinsic chemical reactivity alone controls the particle reaction rate. In other words, the intrinsic kinetics obtained from the chemical reactivity control (regime I) should reflect solely the rates of the chemical reactions. The intrinsic kinetic rates should not depend on the size of the particle and diffusion. This implies that the intrinsic kinetics can be used for PF (particle size: 75–150 μm) and CFB (particle size: 500–6000 μm). In reality, however, the intrinsic kinetics cannot be obtained for a large particle because it is theoretically obtained under isothermal particle conditions. This means that the intrinsic kinetics cannot be obtained because of the difference between the center temperature and the surface temperature caused by the size of the particle. In the previous investigations [25,26], coal char ignition started at a spot on the surface and grew to encompass the entire particle within a few seconds. However, most of the previous research [16,27–32] has used the energy balance method, assuming that the particle ignites uniformly over its surface; it is assumed that the char particle undergoes an isothermal reaction. Therefore, a model is needed that can obtain the intrinsic kinetics for a large char particle using the theory of particle internal conduction. Under experimental conditions, the observed combustion rate is lower than the maximum rate controlled by the intrinsic kinetics because the char combustion and the particle layer can be influenced by concentration gradients arising from oxygen diffusion processes [33]. In order to obtain the intrinsic reaction kinetics from the char combustion