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Soot loading, temperature and size of single coal particle envelope flames in conventional- and oxy-combustion conditions (O_2/N_2 and O_2/CO_2)

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ABSTRACT

A fundamental laboratory study on the volatile-phase combustion of pulverized coal was conducted at the *particle level*. A primary goal has been to simultaneously assess soot volume fraction, f_v, and soot temperature, T, in the diffusion flame (soot mantle) forming around a single burning bituminous coal particle, upon ignition of its volatile matter. This assessment was conducted with emission-based pyrometric methods. A secondary goal of this study has been to compare these radiative parameters, f_v and T, in conventional air- and in simulated dry oxy-fuel combustion. Both f_v, and T measurements were spatially averaged in the flame but temporally resolved throughout the combustion history of single particles. In addition, the size of the volatile envelope flames was assessed both pyrometrically and cinematographically. Combustion of three different bituminous coals took place with various oxygen partial pressures in nitrogen and in carbon dioxide background gases. Single particles, 75–90 μm, were injected and burned in a transparent drop-tube furnace (DTF) at laminar-flow atmospheric-pressure and a wall temperature of 1400 K. The free-falling bituminous coal particles heated up and devolatilized, their volatile matter ignited and formed bright envelope flames, often with distinctive soot contrails in the wakes of the flames. The radiative parameters f_v, T and flame size, of these luminous envelope flames were assessed using different emission-based models for the analysis of the three-color pyrometric intensities. The particle envelope flames of all three coals were found to contain comparable to each other soot volume fractions, in the range of 20–90 ppm. At identical furnace gas temperatures and identical O_2 mole fractions, when the background N_2 gas was replaced with CO_2, the particle envelope flames of the bituminous coals were characterized by lower soot volume fractions, lower temperatures and bigger sizes. As the O_2 mole fraction increased in either N_2 or CO_2 background gases, soot volume fractions increased to a maximum and then decreased, temperatures increased monotonically and flame sizes decreased. In CO_2-based combustion, an oxygen mole fraction in the neighborhood of 35% was necessary to elevate the measured flame temperature to match that of conventional air-based combustion; however, the soot volume fraction was lower than that in air-based combustion regardless of the oxygen mole fraction.

1. Introduction

Soot consists of submicron carbonaceous particles which form in the pyrolysis of hydrocarbon fuels. During combustion of solid fuels, such as coal, soot is generated when the devolatilizing volatile matter therein undergoes secondary reactions at high temperatures in the oxygen-deficient environment of a diffusion flame [1]. Soot is beneficial to combustion systems because of its radiative heat transfer effects; but it may also problematic because of its pollution-generating potential, if it is not burned within the flame envelope [1,2] or in its vicinity.

In coal-fired furnaces, contributions to radiative heat transfer stem from burning soot and chars as well as from hot gases. Evidence of the dominant role of soot luminosity in radiation heat transfer of furnace flames has been long standing [3,4]. Volatile matter flames (soot mantles) forming around devolatilizing coal particles account for most of the total radiant flux into the

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Nomenclature

A	total flame area (μm^2)
c_1	0.59552×10^{-4} ($W \mu m^2/Sr$)
c_2	14388 (μm K)
C_i	calibration constant
d	flame projected diameter (μm)
D_p	particle diameter (μm)
D_{soot}	soot diameter (μm)
E_{total}	total emissive power (W)
f_v	soot volume fraction
F_λ	absorption coefficient of soot
$g_i(\lambda)$	pyrometer geometric constant at channel wavelength i
$I_{\lambda b}(\lambda)$	Plank's radiation intensity (5.67×10^{-8} W/m^2 μm)
k	imaginary part of the soot refractive index
l	distance between pyrometer and an object (m)
L	soot path length (μm)
MW_{soot}	soot molecular weight (gr/mol)
n	real part of the soot refractive index
P_{total}	gas total pressure (Pa)
R	universal gas constant 8.314 (J/mol K)
S	soot mole fraction (moles of carbon/mole of gas)
S_i	experimental signal at channel i (V)
s_i	theoretical emissive power at channel i (V)
T	flame (soot) temperature (K)
T_w	furnace wall temperature (K)
Δt	flame burnout time (ms)

Greek symbols

α	constant: 1.39 for visible and 0.95 for infrared wavelengths
ε_{total}	total flame emissivity
$\varepsilon_{\lambda i}$	flame spectral emissivity at channel wavelength i
σ	Stefan–Boltzmann constant (5.67×10^{-8} W/m^2 K^2)
λ	wavelength (μm)
$\Delta\lambda$	wavelength channel width (μm)
Δ_i	least square error
ρ_{soot}	soot density (g/cm^3)

$\gamma_s = D_{ins}/D_f \gamma_p = D_p/D_f$

Subscripts

i	wavelength channels (998, 810, 640 nm)
f	flame
p	particle
ins	instantaneous inside flame
r	reference source

surroundings during the initial stages of coal particle combustion [5,6]. In the early period of devolatilization after an envelope flame has been formed, the surface temperature of a coal particle is low by comparison to the temperature of the volatile matter flame [6], as experimentally illustrated by Timothy et al. [6], Atal and Levendis [7], and numerically calculated by Lau and Niksa [5]. In fact, the latter authors also calculated that in the case of a 70 μm bituminous coal particle (Pittsburgh #8) the radiation from the soot mantle during the devolatilization phase is up to 1000 times greater than the radiant flux from the surface of the devolatilizing coal particle itself. Soot volume fraction has been identified as the geometric feature of soot that affects radiation more than soot particle size and shape [5]. The soot volume fraction in a coal flame, $\boldsymbol{f_v}$, along with its temperature, is a key parameter in determining the radiative heat transfer in a furnace [8,9], as the total radiative power is proportional to $\varepsilon_{total} A\sigma T^4$, where: the total emissivity is given by $\varepsilon_{total} = 1 - [1 + k\boldsymbol{f_v}LT/c_2]^{-4}$ [3].

In recent years, oxy-fuel combustion has been considered a viable technology to reduce emissions of pollutants and facilitate capture and sequestration of CO_2 which is a greenhouse gas [10]. Although many aspects of this technology have been studied [10], only few investigations have reported on the soot emissions relevant to oxy-fuel combustion, including Refs. [11–13]. As mentioned earlier, the contributions of soot to the radiative heat transfer of the flame are of paramount importance in a boiler; thus, lower temperatures and decreased luminosity of particle flames under oxy-combustion conditions [14–16], along with lengthier ignition delays [14] can be detrimental. In fact, Morris et al. [11,12] reported that burning bituminous and subbituminous coals in a pilot-scale laboratory combustor under both simulated oxy-fired and actual recycled flue gas conditions generated a lesser amount of soot emissions than combustion of these coals under conventional air conditions. However, Stimpson et al. [13], using a line-of-sight laser extinction method in two large laboratory furnaces, obtained results that were not as clear on the comparison of air- and oxy-coal- combustion. In one combustor, coal firing with 25%O_2/CO_2 generated less soot than firing with air, coal firing with 25–30%O_2/CO_2 generated equal soot to firing with air, and coal firing with 30–35%O_2/CO_2 generated more soot than firing with air. The flow rates and, thus mixing differed in all cases, which the authors said indicated the dominance of flow dynamics in soot generation. In the other combustor, however, air-fired flames produced consistently a higher concentration of soot than the oxy-fired flames. Moreover, combustion in this furnace generated soot volume fractions that were highest in air, followed by oxy/FGR, and then by oxy/CO_2 flames. Stimpson et al. [13] attributed this behavior to physical reasons rather than chemical reasons, as they noticed that oxy-coal flames experienced more liftoff than flames in air, allowing more oxidizer entrainment, leaner combustion and, therefore, less soot.

To investigate the aforementioned somewhat inconclusive results obtained from observations at the *flame level*, in multi-particle pulverized conventional and oxy-combustion [11–13], a basic investigation was performed herein at the *particle level*, to assess the soot volume fraction, f_v, in particle envelope flames. Combustion of single particles of bituminous coals occurred in various O_2/N_2 and O_2/CO_2 atmospheres, under quiescent flow conditions and f_v was deduced using an emission-based method. The investigation of single particles under analogous and well-characterized conditions eliminated many of the complicated phenomena present in real systems allowing an examination of some of the more fundamental processes. Of course, subsequent translation of fundamental research to practical applications would need to be done carefully to account for the phenomena present in a particular situation. This task, however, is beyond the scope of the present work.

Whereas the char combustion phase of coal at the particle level has been already studied extensively, see for instance Refs. [6,7,14–28], reports on the volatile matter combustion phase of coal at the particle level have been of limited number, see Refs. [2,7,14,15,18–20]. Even more limited have been reports on the soot volume fraction in volatile matter flames that surround coal particles. Mc Lean et al. [18] observed that during combustion of bituminous coal particles a condensed soot-like phase forms by released volatile matter, which is subsequently either consumed under locally oxidizing conditions or persists throughout the furnace under locally reducing conditions. Seeker et al. [19] observed trails of soot during volatile combustion of bituminous particles bigger than