



A numerical investigation of the effect of flue gas recirculation on the evolution of ultra-fine ash particles during pulverized coal char combustion



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ABSTRACT

Flue gas recirculation (FGR) has been widely employed for NO_x reduction. However, because PM removal in flue gas is never 100% efficient, a gradually accumulated particle concentration (particularly the ultra-fine particles, smaller than 1.0 μm in aerodynamic diameter) may occur in the furnace, affecting PM emission and power plant operation. Experimental measurements cannot provide detailed information on the evolution of ultra-fine particle (including formation, and time-dependent growth and number size distribution). In this work, an improvement over our previous effort to model ultrafine particle formation during coal char combustion is developed and applied to elucidate the effects of various FGR parameters on ultrafine particle formation, including FGR ratio, dust removal efficiency, and particle size distribution in FGR. Model results show that FGR without recirculated particles results in the nucleation of fewer but larger particles. However, the presence of diluting FGR gases results in a low particle collision frequency and consequently yields small particle size and high number density after coalescence. The recirculated particles not only provide available surface area for condensable mineral vapors but also take part in coalescence via collision; consequently, larger particles are formed at a lower overall number density. The ultra-fine particle size first increases slowly with an increase in dust removal efficiency, but then drops rapidly after reaching a peak; An increase in recirculated particle size and a decrease in FGR results in increased particle size. For all cases, the particle number density shows opposite trends to particle size. Overall, the results derived here provide meaningful guidelines for practical application.

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1. Introduction

In conventional air combustion, the recycled hot flue gas mixed into combustion air facilitates fuel vaporization [1] and air-staged firing [2,3], both of which improve NO_x reduction. Therefore, flue gas recirculation (FGR) has been widely adopted in coal power plants for NO_x reduction. In addition, FGR reduces the requisite excess air and reduces the SO₂ and particulate matter (PM) emissions – particularly the PM, which can be reduced by greater than 33%, depending on where the recycled flue gas is extracted downstream of the boiler [4].

However, because PM capture in flue gas is never 100% efficient, higher concentrations of these particles (particularly for ultra-fine

particles, smaller than 1.0 μm in aerodynamic diameter) may occur, potentially resulting in high PM emission [5] and serious slagging and corrosion [6]. Although the ultra-fine ash particles account for only 0.2–2.2% of the total fly ash mass from pulverized coal combustion in boilers, they are difficult to be captured by dust separators and make up nearly all of the particles that discharge into the atmosphere [7]. Moreover, the ultra-fine ash particles may contain a variety of trace toxic compounds and can penetrate deeply into the lungs and consequently cause various diseases [8–11]. Fly ash enriched in ultra-fine particles is of increasing environmental concern [12]. According to the most recent Chinese air pollution emission standard for thermal power plants, PM emissions from coal power plant are limited to 30 mg/Nm³, and in some key regions they are restricted to 20 mg/Nm³ [13] and have been suggested to strive to attain a limit of 10 mg/Nm³ [14].

Recently, Morris et al. [5] studied the effects of various cleanup options prior to FGR on ash aerosol formation characteristics in

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Nomenclature

Symbols

C_{ar}	carbon content in coal as received basis
d, d'	diameter (cm)
D, D'	diffusion coefficient (cm^2/s)
EOC	excess oxygen coefficient
f	number fraction
F	mass fraction
FCO_2	fraction of CO_2
FO_2	oxygen content in combustion gas
FGR	Flue gas recirculation
H_{ar}	hydrogen content in coal as received basis
I	rate of nucleation ($\text{N}/\text{cm}^3/\text{s}$)
k_B	Boltzmann constant, $1.38 \times 10^{-23} \text{J/K}$ cohesion factor (cm^3/s)
K	reaction equilibrium constant
$Kn,$	Knudsen number
m	mass (g)
M	molar mass (g/mol)
M_{ar}	moisture content in coal as received basis
MO_n	metal oxides
MO_{n-1}	metal suboxides and metals particle number concentration (N/cm^3)
N	Inclusion number in one coal char particle
n	the gaseous number condensed on specified sized particle
N_{ar}	nitrogen content in coal as received basis
O_{ar}	oxygen content in coal as received basis
q	burning rate ($\text{g}/\text{cm}^2/\text{s}$)
P	pressure (atm, Pa)
r	radius (cm)
\bar{r}	average radius of the pore channel (cm)
R	ideal gas constant ($8.314 \text{J}/\text{mol}/\text{K}$)
S	extent of saturation
S_{ar}	sulfur content in coal as received basis
S_g	internal specific surface area (cm^2/g_C)
t	coalescence time (s)
T	temperature (K) volume (cm^3/s)
V	vaporization rate (mol/s)
Y	ash content
ΔG_v	change in Gibbs' free energy for droplet formation per unit volume vaporization rate (mol/s)
V	ash or carbon volume in char core (cm^3) gas volume (Nm^3/kg coal)

Greek alphabet

α_c	condensation coefficient
ρ	mass density (g/cm^3)
η, η'	effectiveness factor
θ	porosity
θ'	volume fraction of inclusion in char
π	Pi, 3.1415926
v	molecular volume (cm^3)
τ	tortuosity
σ	surface tension of liquid
σ_c	constriction factor

Subscript

0	initial, standard stage
a	ash
a,f	porous ash
c	carbon
cc	char core

CO_2	carbon dioxide
crit	critical
cond	condensation
eff	effective
eq	equilibrium
cg-theory	theoretical combustion gas requirement
fg	flue gas
g	gaseous
H_2O	steam
i, j	serial numbers of different sized particles
int	intrinsic
l	liquid
M	metal sub-oxide or metal
MO	metal oxide or inclusion
O_2	molecular oxygen
O_2 -theory	theoretical oxygen requirement
s	surface, solid
tot	total
vap	vaporization
Superscript	
eq	equilibrium
s, s'	surface

a 37 kW down-fired pilot-scale combustor, including (a) no clean up whatsoever, (b) removal of particles alone, (c) removal of particles and water vapor, and (d) removal of sulfur, water and particles. They reported that the ultra-fine particle concentrations increase in the boiler despite flue gas treatment with fabric filters, which may fail to remove the nano-particles. Furthermore, Jiao et al. [12] studied the ash partition properties with FGR and found that most fine ash particles with a primary size down to $1.0 \mu\text{m}$ or less exhibit fractal structures because of homogeneous condensation of metallic vapors and/or their heterogeneous condensation on pre-existing fine mineral grains, followed by agglomeration.

During coal char combustion, the continuous vaporization of inorganic components from the char matrix, and subsequent homogeneous nucleation, heterogeneous condensation, coagulation, and coalescence play dominant roles in ultra-fine ash particle formation [11,16]. As shown in Fig. 1 [6,15], during coal combustion, the organo-metallic compounds (such as Cr and V) first vaporize during the devolatilization stage, and then volatile trace elements (Hg, As, and Se etc.) and alkali and alkali-earth elements (K, Na, Mg, and Ca) vaporize and diffuse out of the char matrix during the char burning stage. Meanwhile, refractory elements (Si, Al, and Fe) partially volatilize, but most of their element mass remains with the char particle. With an increasing accumulation of volatilized metals in the gas flow during char burnout, inorganic particle nucleation, condensation, coagulation, and coalescence occur, forming ultra-fine ash particles [6,11,15]. Except for the fraction of ash forming ultra-fine ash particles, most ash compounds in the raw coal remain in the residual fly ash, and the fraction of ash remaining in trace vapors (including Hg, As, Se, etc.) is negligible.

Substantial research has been conducted on the major factors that affect the formation of ultra-fine ash particles, such as combustion temperature and gas atmosphere, as well as coal type [10,11,17–21]. Elevated temperatures enhance mineral vaporization, and subsequent condensation, coagulation and coalescence [11,18], whereas a low oxygen concentration reduces the formation of ultra-fine ash particles [10]. Meanwhile, the use of lower rank coals and the presence of a locally reducing atmosphere in the char matrix can enhance the formation of ultra-fine ash particles because of higher alkali metal content in the char, a higher char combustion temperature (associated with higher char reactivity), and the impact of CO on mineral vaporization [11,21].

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