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Ultra-fine particulate matters (PMs) formation during air and oxy-coal combustion: Kinetics study

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HIGHLIGHTS

- Ultra-fine PMs formation during coal char combustion is studied using kinetics model.
- Ultra-fine PMs formed in oxy-coal atmosphere have fewer number but larger size.
- Ultra-fine PMs show increasing number density and decreasing size with increasing FGR.
- Oxy-coal combustion advantages PMs removal.
- Elevated Flue gas recirculation ratio disadvantages PMs removal.

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ABSTRACT

Although both air ($21O_2/N_2$) and oxy-coal ($27O_2/CO_2$) combustion are widely adopted in pulverized coal (PC) fired power plants, the formation mechanisms of ultra-fine PMs during PC char combustion under both atmospheres with various flue gas recirculation (FGR) ratios are still unclear. Moreover, conventional experimental measurement devices cannot provide detailed information on the formation and evolution of the size-number of ultra-fine PMs. Therefore, the formation of ultra-fine PMs during PC char combustion under both atmopspheres with and without FGR are studied by a self-developed Char Burning and Particulate Matters Kinetics model (CBPMK). The PC char shows similar burning temperature and thus ash vaporization rate under both atmospheres, whereas the vaporization amount under oxy-coal combustion atmosphere is lower than that under air combustion atmosphere due to the shortened burnout time caused by CO_2 gasification reaction under the former. Consequently, during nucleation and condensation stages (before successive coalescence), both the particle size and number under air combustion atmosphere are higher than those under oxy-coal combustion atmosphere. However, after coalescence, the final particle shows fewer but larger size under oxy-coal combustion atmosphere due to the higher cohesion factor between the smaller sized nucleation particles, which improves particle collision and coalescence. Meanwhile, both mean size and number density of the nucleation particles decrease with increased FGR ratio under both air and oxy-coal combustion atmospheres, however, after coalescence the final PMs show increasing number density and decreasing size. As results, oxy-coal combustion advantages PMs removal through an ash collector, but elevated FGR ratio disadvantages PMs removal. Oxy-coal combustion with low FGR ratio should be recommended in PC thermal and power plants.

1. Introduction

Together with sulfur oxide [1,2] and nitrogen oxide [3–6], particulate matters (PMs) emitted from pulverized coal (PC) combustion power plants play crucial role during the formation of haze and serious environmental pollution [5,7–11]. According to the most recent national air pollution emission standard for thermal power plants in China, PM emissions from coal power plant are limited to 30 mg/Nm^3, and in some key regions they are restricted to 20 mg/Nm^3 [12] and suggested to strive to 10 mg/Nm^3 [13] or 5 mg/Nm^3 in some provincial standards. The PMs capture and removal in flue gas is never 100% efficient during coal combustion, particularly of ultra-fine ash particles (smaller than 1.0 μm in aerodynamic diameter), which constitute most of the particles that discharge into atmosphere directly [14] or accumulate in the furnace through flue gas recirculation (FGR) and result in an increase in number density due to the failure of conventional filters

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Nomenclature

Symbols

d, d'	diameter (cm)
D, D'	diffusion coefficient (cm^2/s)
f	number fraction
FGR	flue gas recirculation
I	nucleation rate ($\#/m^3/s$)
k_B	Boltzmann constant (1.38×10^{-23} J/K)
K	cohesion factor (cm^3/s)reaction equilibrium constant
Kn	Knudsen number
m	mass (g)
MO_n	metal oxides
MO_{n-1}	metal suboxides and metals
n	number of mineral vapor molecule condensed on specified sized particle ($\#/m^3$)
N	inclusion number in one coal char particle; or particle number concentration ($\#/m^3$)
q	burning rate ($g/cm^2/s$)
P	pressure (atm, Pa)
PC	pulverized coal
PMs	particulate matters
r	radius (cm)
R	ideal gas constant (8.314 J/mol/K)
S	saturation degree of the vapors
S_g	internal specific surface area (cm^2/g C)
T	temperature (K)
t	coalescence time (s)
Y	ash content
ΔGv	change in Gibbs' free energy for droplet formation per unit volume
V	vaporization rate (mol/s)

Greek alphabet

α_c	condensation coefficient
ρ	mass density (g/cm^3)
η, η'	effectiveness factor
θ	porosity
ν	molecular volume (cm^3)
σ	surface tension of liquid

Subscript

a	ash
c	carbon
cc	char core
CO	carbon monoxide
CO_2	carbon dioxide
eff	effective
eq	equilibrium
g	gaseous
H_2O	steam
i, j	serial numbers of different sized particles
int	intrinsic kinetics
MO_{n-1}	metal sub-oxide or metal
MO_n	metal oxide or inclusion
O_2	molecular oxygen
s,l	solid, liquid
tot	total

Superscript

eq	equilibrium
s, s′	surface

in removing the particles [10]. The emitted PMs from coal-fired furnace may be enriched with trace toxic compounds, which not only cause atmospheric haze but also penetrate into the lungs, consequently causing a number of diseases [15,16]; the accumulated PMs in furnace accelerate the abrasion of heating-surface and deposition formation on its surface, resulting in reduced boiler efficiency and operational safety [17,18].

Respecting of the formation of ultra-fine PMs during coal/char combustion, numerous experiments and kinetics modeling researches on the forming mechanisms and the effects of combustion temperature and atmospheres as well as coal types and FGR have been well documented. By an experiment conducted in a lab-scale drop-tube furnace, Jiao et al. [19] found that most ultra-fine PMs exhibited fractal structures due to the homogeneous nucleation of metallic vapors and/or their heterogeneous condensation on preexisting fine mineral grains during coal combustion with FGR. During coal char combustion, part of inorganic components (Si, Al, Fe, Ca, Na, etc.) in char matrix undergoes successive vaporization, homogeneous nucleation, heterogeneous condensation, coagulation, and coalescence to form ultra-fine PMs [7,9,20–22]. On basis of the abovementioned formation mechanisms of ultra-fine PMs, we developed a Char Burning and Particulate Matters Kinetics model (CBPMK) [9,22]. The modeling results showed that combustion temperature had a dominating positive effect on mineral vaporization, condensation, coagulation and coalescence, and subsequent ultra-fine PMs formation [21–23]. Consequently, the factors including high oxygen content, low ash content, and small sized char particles, which elevated the local char burning temperature [24], caused more and larger ultra-fine PMs [22,25]. Also, Xu et al. [7] studied the vaporization behavior of silica in wet recycle oxy-coal combustion conditions by modeling and experiments conducted in a high temperature drop tube furnace, where the synthetic chars with SiO_2 inclusions were burned at 1873 K. They found that H_2O in combustion atmosphere significantly enhanced the vaporization of SiO_2 and considerably increased the yield of the ultra-fine PM, and the char burning temperature and gas properties surrounding the mineral inclusions seemed to be the primary influencing factors on SiO_2 vaporization. In addition, low rank coals, which generally cause high combustion temperature, and a locally reducing atmosphere, which improves mineral reduction and subsequent vaporization in the char matrix, can enhance the formation of ultra-fine PMs [21,26].

In recent years, respecting of CO_2 capture and storage (CCS) due to the more and more attention on environment pollution, oxy-coal combustion has been developed rapidly [1,27–29]. Sheng et al. [29] studied the impact of O_2/CO_2 combustion on ash particle formation in a drop tube furnace, and found that O_2/CO_2 combustion significantly affected the size distribution of submicron particles, O_2/CO_2 combustion at the same oxygen concentration shifted the size of the submicron mode center to smaller size and decreased the yield of the submicron particles in comparisons with air combustion. Under O_2/CO_2 atmospheres, the char burning temperature is lower than that under O_2/N_2 atmosphere with the same O_2 content due to endothermic gasification reaction and low diffusion of O_2 in CO_2 [24,30], as a result, the number density of the ultra-fine PMs is higher, and the particle size is larger [22,31]. In addition, Morris et al. [10] studied the effects of various cleanup options prior to FGR on ash aerosol formation in a 37 kW down-fired pilot-scale combustor and found that the ultra-fine particle concentrations increased in the combustor despite flue gas treatment with fabric filters. The experiment results are consistent with the kinetics research using CBPMK, indicating that FGR caused a decrease in PMs size with increasing number density [9].