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Characteristics of fine particulate matter formation during combustion of lignite riched in AAEM (alkali and alkaline earth metals) and sulfur

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ABSTRACT

The high contents of AAEM (alkali and alkaline earth metals) and sulfur in zhundong coal result in more fine particulate matter during coal combustion. In this paper, the emission of fine particulates from zhundong coal riched in AAEM-sulfur and zhunnan coal riched in aluminum-silicon was investigated in a drop tube furnace. Co-combustion of zhundong coal and zhunnan coal was further investigated to study the effect of interaction among different elements in coal on the PM formation. The size distribution, concentration and composition of the particulates were analyzed by low pressure impactor (DLPI) and scanning electron microscopy (SEM). The distribution of sodium, magnesium, calcium, iron and sulfur in fine particulates and their effects on the generation of fine particulates were obtained. The results show that the fine particulates from AAEM-sulfur rich coal combustion mainly consist of sulfates and oxides of AAEM. The amount of fine particulates from burning aluminum-silicon rich coal decreased significantly. The composition of $PM_{0.4}$ and $PM_{0.4+}$ are significantly different. Co-firing of AAEM-sulfur rich coal with aluminum-silicon rich coal has an obviously synergetic effect of reducing the fine particulates emission. The capture of AAEM and iron by aluminum-silicon compounds plays an important role in PM_{10} reduction during blended coal combustion. The change of sulfur content in $PM_{0.4}$ is consistent with the change of AAEM content.

1. Introduction

Coal will still be the main energy resource in china for a long time. Recently a large integrated coalfield was found in Xinjiang with a forecast reservation of 390 billion tons [1,2]. It can be used for more than 100 years at the current coal consumption rate of china and will be the main energy base in the future of china. Zhundong coal is characterized by high volatile, low ash content, good combustion characteristics and low mining costs [2]. It is considered a good thermal coal.

However, the extensive usage of coal has caused serious air pollution [3], especially the current removal efficiency of fine particles in coal-fired power plant is still low [4,5]. The combustion of zhundong coal will generate more fine particulates due to the high content of AAEM (alkali and alkaline earth metals) in raw coal [6]. It is important to study the characteristics of fine particles and control the PM emission during the clean utilization of zhundong coal. There have been many researches on the formation mechanisms of fine particulate matter from coal combustion and the formation of submicron particles is considered to be related to the gasification of inorganic elements in coal [6–10]. Most of the alkali metal in coal was water-soluble especially sodium [11]. The alkali metal is easy to vaporize during coal combustion and will condensate to be fine particulate matter as the flue gas is cooling [8,11]. The formation of larger particles (PM_{1+}) is usually related to the evolution of refractory minerals in coal [10,12,13]. Zeng [14] studied the characteristics of fine particulates from coal firing in air at the combustion temperature of 1173 K, 1273 K, 1373 K and 1573 K. He found an increase of magnesium and calcium with a decrease of sodium and sulfur in the fine particulate matter when increasing the combustion temperature. The reason of PM_1 reduction is considered to be the absorption of sodium by fly ash. Li [6] investigated the fine particles formation during combustion of zhundong coal in a 25 KW down-fired furnace. It was found that the $PM_{0.4}$ is mainly composed of sodium sulfate. With the increase of particle size, aluminosilicates become the main component of fine particulates. Gao [7] collected the ultra-fine particles generated from zhundong coal combustion by the thermophoresis micro-probe from the Hencken burner. The analysis results of TEM show that the amount of sodium and silicon is highest in the ultra-

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fine particulates. The $Na_2O{\cdot}nSiO_2$ is thought to be the precursor of ultrafine particles. In the process of coal combustion, the refractory oxides (magnesium oxide, calcium oxide, silicon oxide, iron oxide) can be reduced to more volatile sub-oxides (silicon monoxide) or simple substances (magnesium, calcium, silica, iron) [6,8]. The volatilization of these refractory oxides also lead to the formation of fine particulates and precursor of PM. So far, the researches on fine particulate matter from coal combustion are mainly focused on alkali metal, especially the contribution of sodium in fine particulate formation [11,15,16]. In the previous researches of our group [1,17], the ash deposited on the heating surface of a 350 MW coal-fired power plant showed a layered-structure and were rich in calcium sulfate. The reaction between calcium oxide and sulfur promotes the formation of the initial layer during the ash deposition process. However, the role of alkaline earth metal on the formation of PM has not been fully studied [18].

In this work, a zhundong lignite riched in AAEM and sulfur were burned in a drop tube furnace to study the formation of PM. The co-firing of zhundong lignite and high content aluminum-silicon zhunnan coal was further carried out to study the characteristics of PM reduction during co-combustion. Through the analysis of the main elements in PM among different combustion condition, the effect of sodium, magnesium, calcium, sulfur and iron on the PM formation was discussed in detail.

2. Experimental

2.1. Coal properties

The two coals used in this work were sampled from a coal-fired power plant in Xinjiang province. Both the two coal were dried first and then ground to less than 100 μm in size. The coal properties are presented in Table 1. The zhundong coal is rich in alkali and alkaline earth metals, especially sodium and calcium. The amount of silicon and aluminum in zhunnan coal is high, which is 44.90% and 17.33%, respectively.

2.2. The drop tube furnace, sampling systems and analysis method

The experiment was carried out in a drop tube furnace which is shown in Fig. 1. The blending ratio of zhundong coal and zhunnan coal is selected as 10:0, 9:1, 7:3, 0:10 which are expressed as ZD, 9:1, 7:3 and 0:10 later in this paper. The drop tube furnace is 1.2 m high with an isothermal zone of 0.6 m heated by a three-stage silicon carbide rods. The primary air and the secondary air are mixing gas of oxygen and nitrogen at a ratio of 2:8. A self-designed micro powder feeder was used to feed the coal. The residence time is about 3 s with an excess air coefficient of 2.5. The fine particulates in flue gas were sampled by a water-cooled sampling tube. A flow of nitrogen was injected into the sampling tube to quench and dilute the flue gas, preventing the subsequent reactions. A PM_{10} cyclone after the sampling tube was used to separate the particulates with aerodynamic diameter larger than 10 μm from the flue gas. The PM_{10} particles were collected by DLPI (Dekati low pressure impactor) into 13 fractions. The combustion temperature for experiment was 1573 K and the flue gas temperature at the fly ash sampling position was 873 K. Each case was repeated three times to ensure the reliability of the experimental data.

Aluminum foil coated with Apiezon L resin was used to sampling PM_{10} for analysis. The mass of particulate matter on the foil was obtained by a precision electronic balance (Sartorius M2P, 0.001 mg). The particulates on the foil was analysed by scanning electron microscopy equipped with an energy dispersive spectrometer (SEM-eds, JSM-6390A). The main considered elements included sodium, magnesium, potassium, calcium, aluminum, silicon, sulfur and iron.

Table 1
Typical properties of coal and ash used in this study (wt%).

Coal	Proximate analysis				Ultimate analysis				
	M_{ad}	A_d	V_{daf}	FC_{daf}	C_{ar}	H_{ar}	O_{ar}	N_{ar}	$S_{t,ar}$
zhundong	14.36	8.50	30.86	69.14	53.83	2.38	10.61	0.47	0.33
zhunnan	8.88	17.58	36.79	63.21	56.52	3.21	10.60	0.69	0.44
	Ash compositions								
	Fe_2O_3	Al_2O_3	CaO	MgO	TiO_2	SiO_2	SO_3	K_2O	Na_2O
zhundong	8.02	7.68	40.69	6.6	0.65	17.38	11.32	0.55	6.07
zhunnan	9.01	17.33	13.36	5.04	0.71	44.90	6.47	1.26	1.34

M: moisture, A: ash, V: volatile, FC: fixed carbon.

3. Results and discussion

3.1. Mass-particle size distribution of PM_{10}

The mass-particle size distributions of ZD, 9:1, 7:3, ZN are presented in Fig. 2. The y axis represents the ability of fine particulate matter generated from coal combustion and is scaled by 'mg/g coal'. The x axis represents the aerodynamic diameter of particulate matter and is in the form of logarithmic coordinate.

All of the four curves present a bimodal distribution with the peak of fine model locates at around 0.16 μm and the peak of coarse model locates at around 2.5 μm. The fine model of ZN is not clear because the amount of fine particulates generated from ZN is little. There is a transition point locates at around 0.4 μm between the fine and coarse model. When ZD coal is burned separately, the amount of PM_{10} is highest and the submicron particles were significantly higher than that from ZN coal combustion. The submicron particle formation is mainly relates to the nucleation and condensation process of gasified species. Alkali and alkaline earth metals are thought to be the main components of submicron particles [6,11,19]. When burning ZD coal, the alkali metals which are easy to gasify in coal particles will volatilize into the gas phase. The alkaline earth metals (calcium and magnesium) can be reduced to more volatile matter under the reducing atmosphere of char combustion [8]. Meanwhile, a small part of the refractory metals such as iron and silicon will also gasify under the reducing atmosphere of char particles [6,8]. These mineral vapors will react with oxygen when diffusing to the ambient environment from char. The decrease of oxygen partial pressure results in the increase of mineral vapor partial pressure, which leads to the ultrafine particulates formation. These aerosols are the precursors of submicron particles. In the process of devolatilization, volatile combustion and char burnout, the char particles can be expanded and broken to generate particles much larger than that from the process of gasification-nucleation/condensation [10,20]. The components of these particles are consistent with the mineral matter in coal particles [10]. In addition, the mineral matter melting in high temperature of coal combustion can grow up to form particles larger than 1 μm by coalescence [20]. Some researchers think the salt (sulfate and chloride) and oxide of alkali metal are the main component of fine particulates from coal combustion, especially for coal in zhundong coalfield riched in alkali and alkaline earth metals [1,6]. The content of sodium in ZD coal is 3.83 mg/g coal, which is more than twice as much as it in ZN coal for 1.75 mg/g coal. However, the submicron particles from ZD coal combustion are much more than that from ZN coal combustion. This indicates that there are some other factors affecting the formation of fine particulate matter. The content of silicon and aluminum in ZN coal is 6 times of that in ZD coal. The silicon-aluminum species in coal generally form PM_{10+} during coal combustion. The high content silicon-aluminum mineral in ZN coal has an obvious absorption of alkali metal aerosol, resulting in significantly less submicron particles of ZN coal [21].

In order to investigate the effect of co-combustion on the formation of $PM_{0.4}$ and PM_{10}, the calculated value (particles concentration is calculated assuming that there are no interactions among the mineral