EXPERIMENTAL STUDY ON PRODUCTION AND EMISSION CHARACTERISTICS OF $PM_{2.5}$ FROM INDUSTRIAL FLUIDIZED BED BOILERS

by

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Original scientific paper
https://doi.org/10.2298/TSCI190828001Z

Particle size distributions, concentrations, morphological characteristics, and elemental compositions of eight fluidized bed boilers with different capacities and different dust collectors were determined experimentally. The $PM_{2.5}$ particle concentration and mass concentration were monitored in real-time before and after the boiler dust collector by electric low pressure impactor, and the physical and chemical properties of $PM_{2.5}$ were analyzed by membrane sampling. We found that the $PM_{2.5}$ particle concentration produced by industrial fluidized bed boilers displayed bimodal distributions, peaking at 0.2 μm and 0.76 μm, the formed mechanism of these two parts particles is vaporization-condensation of mineral matter and residual ash particles and the adsorbent wear or tear. Mass concentration exhibits a single peak characteristic with a peak at 0.12 μm. The removal efficiency for $PM_{2.5}$ of dust collectors varies with different dust removal mechanisms. The electrostatic precipitator and bag filter have high dust removal efficiency, and the water film dust collector has low dust removal efficiency. The normal operation of the bag filter has a great influence on the dust removal efficiency. The physical and chemical properties of $PM_{2.5}$ showed that the single-particle morphology was mainly composed of irregular particles, containing a small amount of solid spherical particles and more agglomerates. The content of Si and Al in $PM_{2.5}$ elemental analysis is the highest, which decreases after a dust collector. Some fluidized bed boilers use desulfurization in the furnace, which has great influence on the mass concentration of Ca and S elements, and the lowest Hg content in trace elements, about a few ppm.

Keywords: *industrial fluidized bed boiler, $PM_{2.5}$, concentration, dust removal efficiency, physical and chemical properties*

Introduction

In recent years, smog weather has severely affected the lives of residents. The $PM_{2.5}$ (the particle aerodynamic diameter less than 2.5 μm) that can enter the lungs is a significant pollutant. The $PM_{2.5}$ can float in the air for a long time due to its small gravity and strong fluidity with air-flow. The $PM_{2.5}$ has a large specific surface area and is easy to carry disease-treating bacteria, which has severely affected the health of residents [1-3].

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Coal combustion in source analysis is the primary source of $PM_{2.5}$, and industrial boilers are numerous in China. Fluidized bed boilers have a growing proportion of industrial boilers due to their broad applicability and high economic efficiency. It is of considerable significance to research the characteristics of bed and boiler $PM_{2.5}$ production and discharge [4-6].

Qu *et al*. [7] researched the impact of O_2 concentration and combustion temperature on emission and chemical composition of $PM_{2.5}$ on a 50 KW CFB (circulating fluidized bed). They found that enhancing the oxygen concentration and the combustion temperature increased the emission of $PM_{2.5}$, and the emission of $PM_{2.5}$ appeared in two modes, one is the submicron around 0.2 μm formed through the vaporization and condensation mechanism, the other around 2.0 μm formed through fragmentation and coalescence mechanism. Goodarzi [8] used DEM/EDX to find that most of the particulate matter emitted is spherical, $PM_{>10}$ particles contain hollow microspheres, residual carbon, and polygonal quartz. The PM_{10} particles contain solid spheres, hollow microspheres, gypsum crystal needles, *etc*. Using INAA and ICP-MS to test the content of elements in the particles, $PM_{2.5}$ particles are mostly composed of Si-Al salt solid sphere particles enriched with Ba, Ca, and Fe on the surface; most elements such as Al, Ca, and Fe can be found in the $PM_{>10}$ and PM_{10} particle size segments, but Cd, Cu, Mo, and Ti can only be detected in $PM_{2.5}$ particles.

Domestic and foreign scholars have carried out research on pulverized coal boilers, and there are few studies on fluidized bed boilers, especially the lack of industrial fluidized bed boiler $PM_{2.5}$ generation and emission system analysis. The characteristics of $PM_{2.5}$ particle number and mass concentration real-time monitoring and sampling analysis from different fluidized bed boilers and different dust collectors have guiding significance for the introduction of industrial fluidized bed boiler fine particulate matter emission limits.

Test section

Introduction to the boiler

In this paper, eight fluidized bed boilers with different capacities were selected for the continuous measurement of $PM_{2.5}$ mass concentration and particle concentration before and after the precipitator and particle sampling, including seven CFB boilers and one bubbling bed. The capacity of boilers which the smallest is a 20 t/h steam boiler and the largest is a 116 MW hot water boiler. The dust collector includes old-fashioned mechanical dust removal and new static electricity and bag filter. Since the fluidized bed boiler generally adds additives such as limestone to the fuel, the SO_2 emission concentration can be reduced through desulfurization in the furnace. The test objects in this paper are not equipped with a desulfurization tower. The sampling point's layout of this experiment is shown in fig. 1.

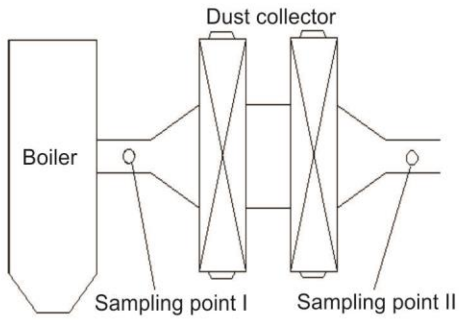

Figure 1. Schematic of the industrial fluidized bed boilers

Sampling and analysis

This test used the electric low-pressure impactor produced by Dekati of Finland to measure $PM_{2.5}$ in the flue gas continuously, and used a two-Stage dilution system with clean air to dilute and cool the high temperature flue gas to atmospheric temperature. The process of cooling, diluting, and coagulating the high temperature flue gas generated by the combustion source to the atmosphere. The two-stage dilution sampling system is shown in fig. 2.

A particle sampler used to sample the $PM_{2.5}$ constant current in the flue gas after the diluter. The sampling membrane contains a polycarbonate membrane and a teflon membrane. The physicochemical characteristics of single-particle morphology, significant elements, and trace elements were analyzed by SEM, XRF, ICP for different samples.

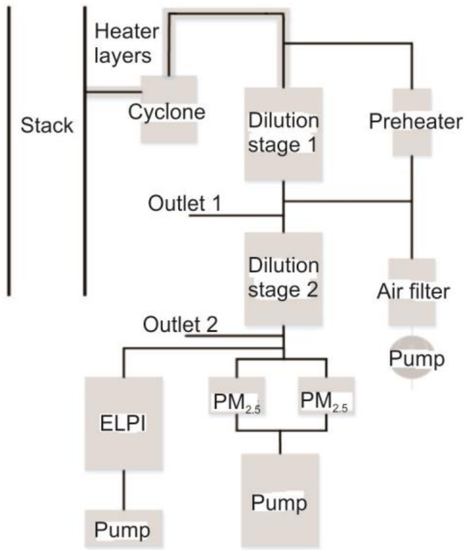

Figure 2. The two-stage dilution sampling system

Results and discussion

The $PM_{2.5}$ particle number concentration and mass concentration emission characteristics from the outlet of boilers

Figure 3 shows the $PM_{2.5}$ particle number concentration distribution from the outlet of boilers. We find that the $PM_{2.5}$ particle concentration produced by the boilers display a bimodal characteristic. The first peak is mainly found at 0.12~0.2 μm, and the second peak is not evident at 0.76 μm. The particle number concentration do not change much with the increase of particle size, and the concentration of particles generated by the industrial fluidized bed boiler is relatively average. Yu *et al*. [9] divided the particles formed by the combustion of pulverized coal into three modes: ultra-fine mode (particle size below 0.3 μm), intermediate mode (particle size of 0.3~3 μm), and coarse mode (particle size of 3 μm above), $PM_{0.12}$ is an ultrafine mode particle, mainly formed by gasification-condensation and nucleation agglomeration mechanism. $PM_{0.3\text{-}2.5}$ is an intermediate mode particle, mainly composed of residual ash particles, and through minerals. Mechanisms such as nucleation agglomeration and wear and tear of the adsorbent are formed.

The boiler 6 is a bubbling bed, and the number of particles at less than 0.12 μm is significantly higher than that of other CFB boilers, which is consistent with Lind's research results [10]. It is considered that the combustion temperature of a bubbling bed is generally 150 K higher than that of the circulation fluidized bed. The higher the temperature, the more ultra-micron particles are broken in the furnace. Some of the ultra-micron particles are entrained by the bubbles to a high position. When the bubbles are broken, they are directly discharged into the flue.

Figure 4 shows the $PM_{2.5}$ mass concentration distribution from the boiler outlet. It can be seen from fig. 4 that the mass concentration of $PM_{2.5}$ produced by the industrial fluidized bed boilers display a unimodal characteristic, and the peak appears at 0.12 μm, which is related to the higher concentration of the particles here. Although the number concentration of the particles decreases slowly after the particle diameter is larger than 0.12 μm, the mass concentration shows an increasing characteristic, indicating that the mass of single particles increases rapidly as the particle size increases, and the mass concentration of $PM_{2.5}$ particles mainly depends on the mass concentration of large particles. The boiler 6# mass concentration is higher than other boilers and the high particle concentration.

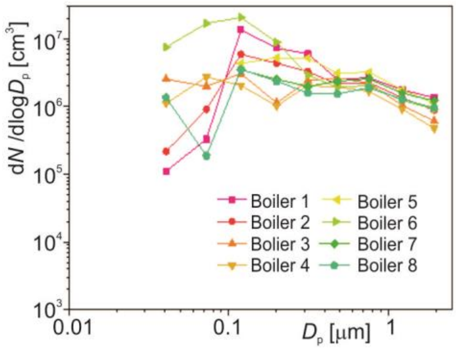

Figure 3. The number concentrations from the outlet of boilers

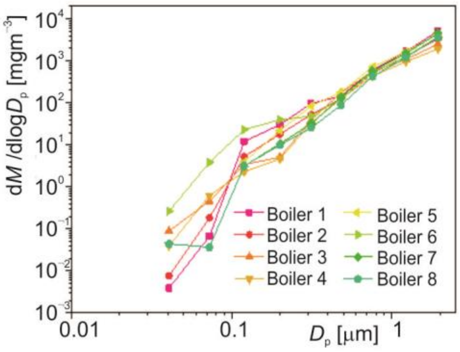

Figure 4. The mass concentrations from the outlet of boilers

The $PM_{2.5}$ particle number concentration and mass concentration emission characteristics after the dust collector

Figure 5 shows the particle concentration distribution of the $PM_{2.5}$ at the outlet of the dust collectors. It can be seen from fig. 5 that the particle concentration of each boiler $PM_{2.5}$ after the dust collector device is significantly lower than that before dust collector, and the particle size distribution of most boilers displays a double peak characteristic. The first peak appears at 0.12~0.3 μm, and the second peak appears at 0.76 μm for some boilers. The peak maybe caused by the low dust removal efficiency and high number concentration from the outlet of boilers at this size range.

Figure 6 is a mass concentration distribution of the $PM_{2.5}$ at the outlet of the dust collectors. It can be seen from fig. 6 that the mass concentration of $PM_{2.5}$ particles in the dust removal device still increases with the increase of the particle size. The mass concentration distributions showing no peak of most boilers, but a less visible peak at around 0.3 μm in boiler 2, which is related to the larger concentration of the particles here. The mass of the single particles is large, and although the concentration of the particles after the peak is decreased, the mass concentration is still increasing.

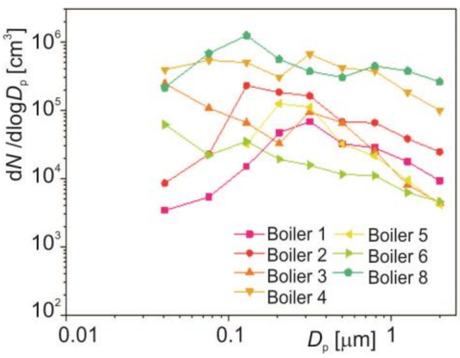

Figure 5. The number concentrations in the outlet of dust collectors

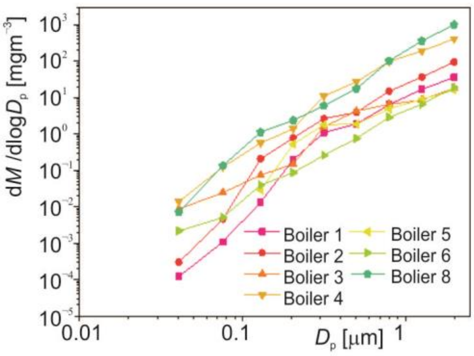

Figure 6. The mass concentrations in the outlet of dust collectors

Analysis of dust removal efficiency of different dust collectors

Table 1 shows the total particle number concentration and total mass concentration of $PM_{2.5}$ after industrial fluidized bed boilers and dust removal devices. The dust removal efficiency of each dust collector to $PM_{2.5}$ particle number concentration and mass concentration is shown in fig. 7.

Table 1. The total number and mass concentrations of boilers

Boiler	Boiler outlet particle number concentration [number cm^{-3}]	Boiler outlet particle number concentration [mgm^{-3}]	Particle number concentration after dust removal [number cm^{-3}]	Mass concentration after dust removal [mgm^{-3}]
1	7357755	1466.64	44861	11.923
2	4397556	1021.336	165501	29.173
3	3826918	1380.354	153423	12.743
4	2952178	1186.076	740108	237.173
5	4827473	1329.435	67181	6.583
6	14054994	2033.016	44392	10.2
7	3195049	1304.38	–	–
8	3241401	1028.11	840731	282.83

The concentration of $PM_{2.5}$ produced by each boiler is several million particles per cubic centimeter, the mass concentration is generally more than one thousand milligrams per cubic meter, and the concentration of boiler 6 is up to 14 million particles per cubic centimeter. The mass concentration reaches more than 2000 milligrams per cubic meter, which is related to the boiler 6 as a bubbling bed. Because bubbling bed boiler do not equipped with a cyclone separator, the fine particles entrained by the flue gas directly enter the flue. Figure 3 showing that the particle number concentration of boiler 6 is significantly higher than other CFB boilers when the particle size is less than 0.12 μm. After the dust collectors, the particle concentration of $PM_{2.5}$ is tens of thousands to hundreds of thousands per cubic centimeter, the mass concentration is generally about ten milligrams per cubic meter, and the bad dust removal efficiency can reach more than 200 milligrams per cubic meter.

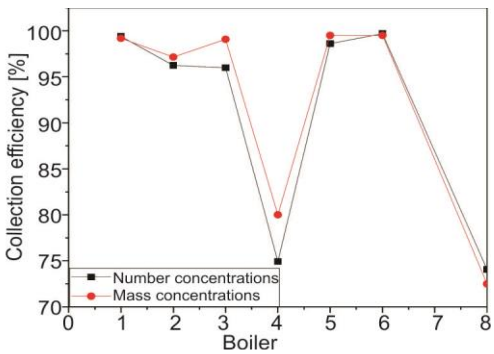

Figure 7. The collection efficiency of number concentrations and mass concentrations

It can be seen from fig. 7 that the dust removal efficiency of the $PM_{2.5}$ particles is different due to the different dust removal mechanisms of different dust removal devices [11]. Boilers 1 and 2 adopt electrostatic precipitators, boilers 3~6 adopt bag filter, boiler 8 adopts water film dust collector. We can find that the boiler 1, 2, 3, 5, and 6 dust collectors have relatively high removal efficiency of $PM_{2.5}$ particle, and the removal efficiency of boiler 5 reaches 99.7%. The boiler 4 dust removal equipment is the bag filter, but the dust removal efficiency of the $PM_{2.5}$ particle concentration is 75%. It may be caused by the damage of the bag in the dust collector. Besides, the dust formed on the surface of the filter bag in the bag filter increases the collision and interception effect between the particles, can capture the particles with smaller particle size, and increases the dust removal efficiency of the bag filter [12]. The lowest dust removal efficiency is the boiler 8 with mechanical dust removal equipment, and the dust removal efficiency of $PM_{2.5}$ is only 72%.

Analysis of the formation of $PM_{2.5}$ particles from the outlet of boilers

The combustion temperature of the fluidized bed boiler is about 1200 K, and the combustion temperature of the coal particle can reach 1700 K. The main gasification in coal is the inorganic element which is connected with the organic functional group in coal in the ion exchange state, such as Na, K, Ca, Mg, and some alkali metals in minerals. These gas-phase elements reach a saturated state with homogeneous temperature and homogeneous nucleation and homogeneous condensation. The particles formed by nucleation are usually tiny, generally from 10 to 100 nm in diameter, and submicron particles are formed by heterogeneous condensation, coagulation or agglomeration.

For the ultra-micron particles, the ash formed by the combustion of internal and external coke is mainly produced. In the combustion process, the crushing of coke has a significant influence on the particle size of the fly ash formed by the mineral particles, for a brittle coal type. Only one mineral particle can lead to a fragment, that is, a mineral particle can produce a fly ash particle, and if it is not broken during the combustion process, all the mineral particles will form a large particle.

Due to the continuous entrainment of particles in the bubbling bed, the transverse particles continuously replenish the space where the particles are removed, causing the horizontal and vertical mixing of the particles to be more intense, while the fluidization speed in the CFB is higher. The returned material from cyclone separator and the secondary wind increases the lateral scouring of the particles, and the fine particles formed by a large number of nucleation are trapped by large particles such as bed materials and additives so that the number of submicron particles is greatly reduced. Since fluidized bed boilers usually use desulfurization in the furnace, a certain proportion of limestone is added to the fuel, and these additive particles have a large particle size, which provides an attachable place for fine particles formed by homogeneous condensation. Qu [13] found that the addition of limestone additives reduces the chance and amount of submicron particle formation.

Morphology of $PM_{2.5}$ particles before and after industrial fluidized bed boiler dust collector

In this study, the $PM_{2.5}$ particles were collected by the polycarbonate membrane at the measuring point before and after the dust collectors, and the morphology was analyzed by SEM. The particle morphology of the industrial fluidized bed boilers from inlet of dust collectors is shown in fig.8, which from outlet of dust collectors is shown in fig. 9. The morphological characteristics of $PM_{2.5}$ from inlet and outlet of the industrial fluidized bed boiler dust collector are unchanged, mainly irregular particles. The combustion temperature of the fluidized bed boiler is generally about 1200 K, and the combustion temperature is relatively low. The melting point of most minerals isn't reached so that the minerals are not dissolved or partially melted to produce a large number of irregular particles, which is different from the pulverized coal boilers [14]. It can also be seen from the figure that the particles collide with each other, large particles collide with some ultrafine particles, and a large number of ultra-micron particles collide and form a chain-like soot polymer, some of the particles appear solid spherical. Due to frequent collision of particles in the furnace, the surface of the solid body particles is attached with a large amount of ultra-micron particles.

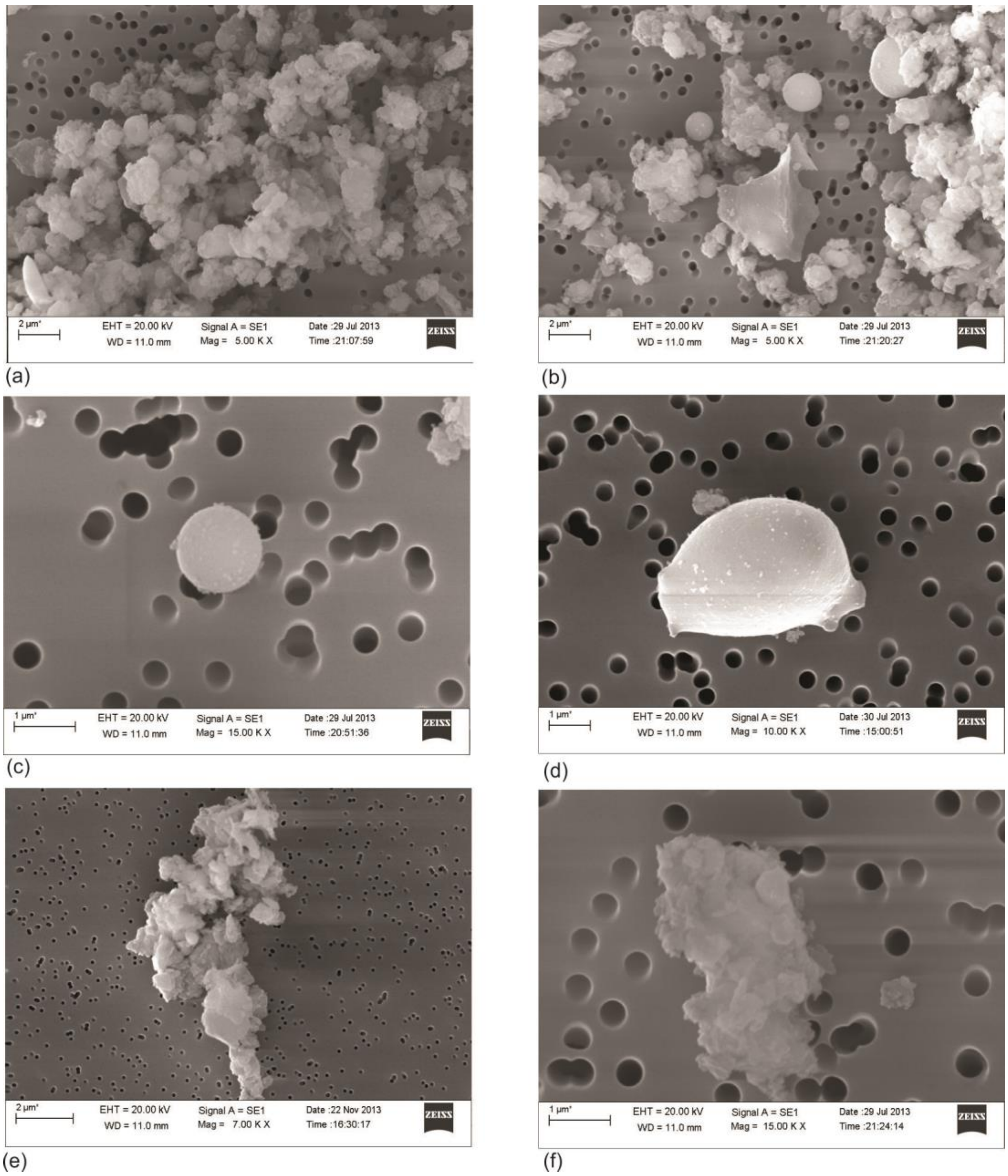

Figure 8. The $PM_{2.5}$ morphology from inlet of dust collectors; *(a) chain-like soot polymer, (b) irregular particles, (c) solid spherical, (d) partially melted particle, (e) irregular particles, (f) irregular particles*

The $PM_{2.5}$ element composition before and after industrial fluidized bed boiler dust collectors

The $PM_{2.5}$ was sampled at the measuring point position from inlet and outletof dust collectors in 6 boilers by teflon membrane. The contents of Na, Mg, Al, Si, S, K, Ca, Fe, and Zn were analyzed by an X-ray fluorescence spectrometer (XRF). The contents of Hg, As, Se, and Pb were analyzed by inductively coupled plasma atomic emission spectrometry (ICP-AES). We calculate the arithmetic mean of the various elements, and the sample element content before and after the dust collectors is shown in fig. 10.

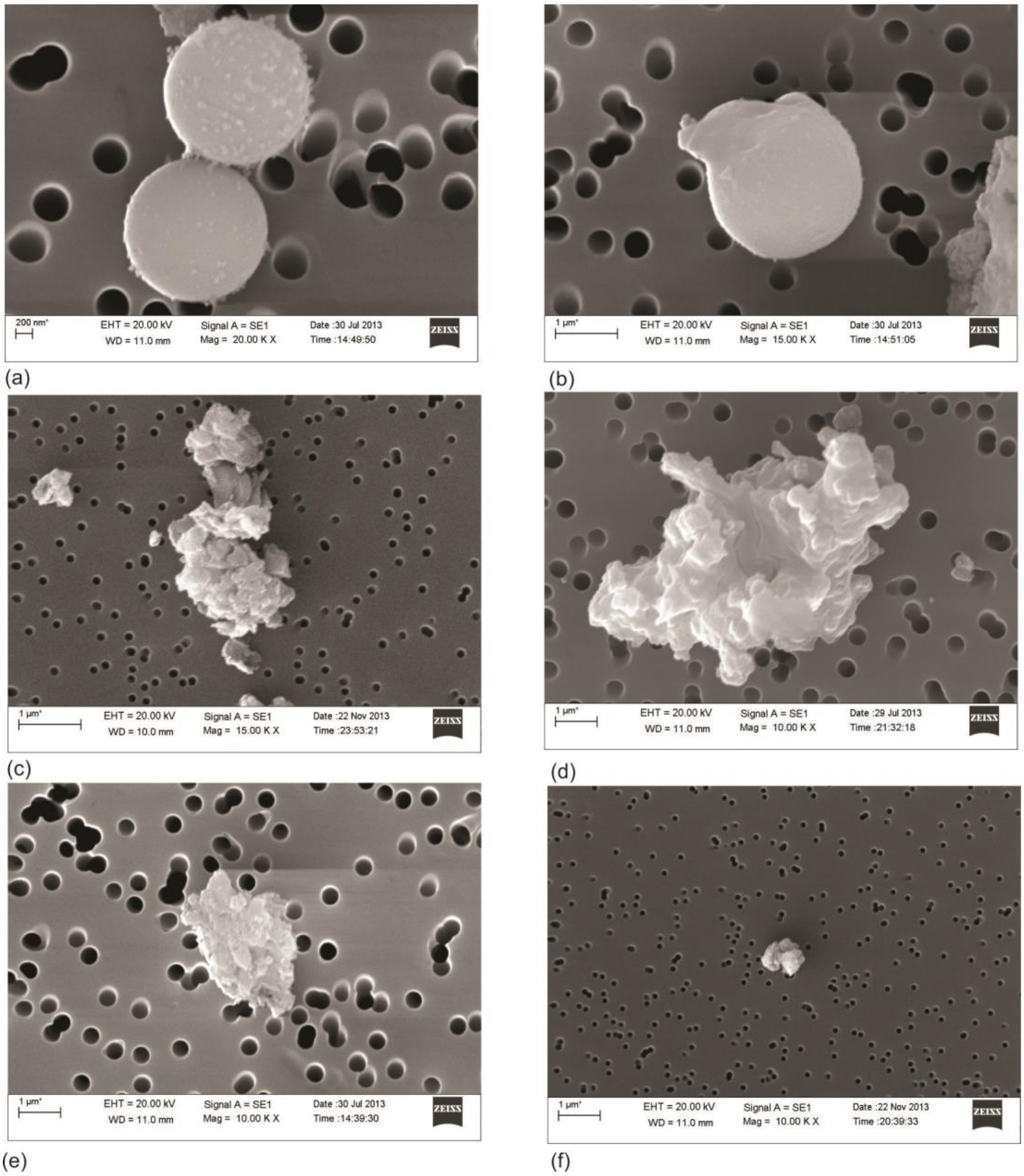

Figure 9. The $PM_{2.5}$ morphology from the outlet of dust collectors; *(a) double spheroidal particle, (b) incomplete melting particle, (c) irregular particles, (d) irregular particles, (e) irregular particles, (f) irregular particles*

It can be seen from fig. 10 that the highest content of elements before and after the dust collectors is Si, the mass percentages before and after the dust collectors are 23.83% and

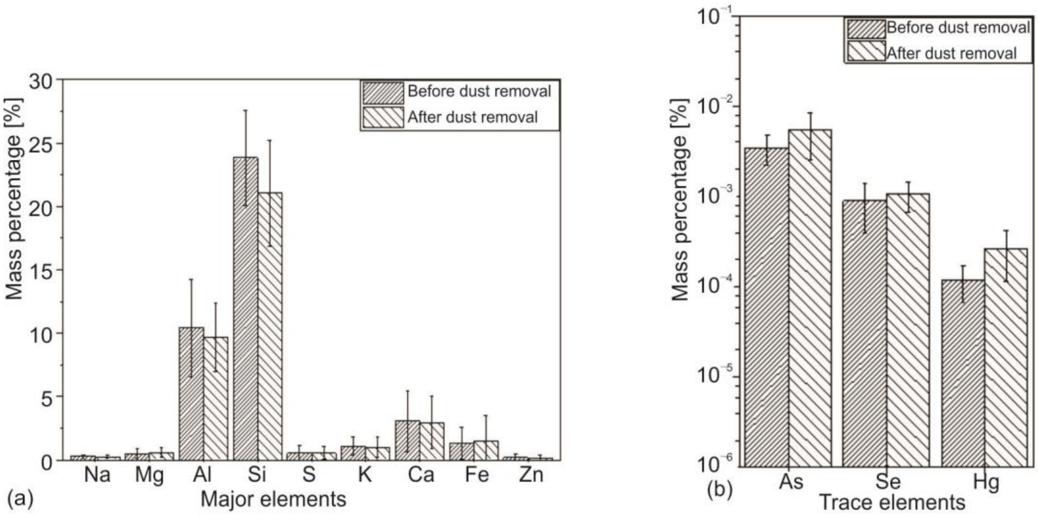

Figure 10. Average contents of representative elements before and after dust collectors

21.07%, respectively, the second is the Al element, the mass percentages are 10.42% and 9.67%, respectively. The melting and boiling points of the Si and Al elements are relatively high, and it may be vaporized in the form of a suboxide under a reducing atmosphere, but the amount of gasification is meager, and the formed aluminosilicate particles are mainly composed of particles having a larger particle size, and the content of Si and Al elements decreased after the dust collectors.. The removal of Ca element is 0.11% lower than that before dust collectors. Because the limestone desulfurization method is used in some boilers and the Ca/S molar ratio is different in the furnace, the Ca content varies significantly between different boilers, and Ca increase the fly ash specific resistance, affecting the effect of electrostatic precipitator on the removal of Ca [15]. The S element content after the dust collectors is low and increased by 0.02% than before dust collectors, because the S element is absorbed by the massive limestone particles and discharged by the large particle size fly ash particles or bottom ash. Due the melting point of S element is low, part of the vapor phase sulfur is condensed on the $PM_{2.5}$ particles during the process of reducing the smoke temperature of the dust collector, increasing the content of S element after dust collectors. Due to the different coal quality components in the combustion, the difference in circulating materials in the bed, the composition of desulfurizer added in different boilers is different and the addition amount mainly depends on the experience of the operating person leads to substantial differences in the elements in the sample, as well as sampling and analysis error factors.

In the fig. 10, we find that the Hg element content is only several ppm, the As element and Pb element content is about 0.01%. Although Hg, As, and Pb are trace elements, they are extremely harmful to the ecological environment and human health [16-18]. China's Ministry of Environmental Protection has added Hg and its compound emission limits 0.03 mg per cubic meter for power station boilers and 0.05 mg per cubic meter for industrial boilers [19, 20]. The emission limits in the standard are the mass of Hg and its compound in the flus gas per cubic meter, but we calculate the mass percentage of Hg element in all elements in this paper. So we cannot compare the experimental data with the emission limits because of the different calculation methods.

Conclusions

In this paper, the $PM_{2.5}$ particle emission characteristics of seven CFB boilers and one bubbling bed boiler with different boiler capacity and different dust collectors are studied before and after the dust collector. The results show that:

- The $PM_{2.5}$ particle concentration produced by the industrial fluidized bed boiler displays a bimodal characteristic. The first peak is at 0.12~0.2 μm, formed by gasification-condensation nodule nucleation, and the second peak is not evident at 0.76 μm and is mainly formed by the mechanism of residual ash particles, nucleation, and agglomeration of minerals, and wear and tear of the adsorbent. Because the temperature in the furnace is higher than that in the circulating fluidized bed, the bubble in the furnace is forcefully broken, and the concentration of fine particles is higher than that of the CFB boiler.
- Different dust collectors have different removal efficiency for $PM_{2.5}$ due to different dust removal mechanisms. The dust removal efficiency of $PM_{2.5}$ for static electricity and bag filter is generally above 95%, and the typical operation bag filter can achieve 99.7% removal efficiency. However, the dust removal efficiency of the non-normal bag filter is only 75%. The dust removal efficiency of the water film dust collector is only 70%, and the old mechanical dust collector cannot meet the environmental protection requirements.
- Industrial fluidized bed boilers due to the low combustion temperature in the furnace, the minerals can only be partially melted. The $PM_{2.5}$ single particles are mainly composed of irregular particles, and some of the particles are solid spherical, and the surface is attached. There are a lot of ultrafine particles.
- The content of Si and Al elements in the $PM_{2.5}$ discharged from industrial fluidized bed boiler is the highest, and the content is decreased after passing through the dust collector. The concentration of Ca and S in different boilers $PM_{2.5}$ varies larger due to the desulfurization method in the furnace. The trace element has the lowest content of Hg, about several ppm, and the content of As and Pb is about 0.01%.

Acknowledgment

This work was financially supported by the project supported by National Key R&D Program of China (2017YFF0209806).

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Paper Submitted: August 28, 2019
Paper Revised: November 11, 2019
Paper Accepted: December 12, 2019