Enrichment Behavior of Sulfur on Ash Particles and Collection Characteristics of Electrostatic Precipitator in Pulverized Coal Combustion System

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Abstract

Physicochemical properties of ash particles and their effects on the collection characteristics of electrostatic precipitator in the pulverized coal combustion system were investigated, focusing on enrichment processes of sulfur on the particle surface. An experimental system composed of one-dimensional laminar flow furnace, cyclone, bag-filter and electrostatic precipitator was used to derive the mutual relations among coal properties, combustion conditions and characteristics of ash particles collected in each device. The following conclusions were obtained.

1) Chemical properties on the particle surface are affected not only by the shape and surface structure of particles but also by particle size, temperature history and coal properties.

2) Soot particulates attached to the small ash particles enhances the enrichment of sulfur on them, and this effect depends on coal properties and combustion conditions.

3) Collection characteristics of ash particles in the electrostatic precipitator largely depend on the enrichment behavior of sulfur in it for the case of high sulfur coal.

1. Introduction

Pulverized coal combustion is now likely to become increasingly important for electrical power generation. Among various pollutants from pulverized coal combustion system, fly ash particles formed from the mineral matter in the coal are one of the most important emissions of environmental concern. In a practical system, almost all fly ash particles are collected mainly in the electrostatic precipitator equipped in the down stream, and the collection performance is largely affected by the electrical properties such as electrical resistivity and dielectric constant of particles, which will depend on the chemical compositions of particle surface. Further, some of hazardous elements such as As, Se, Pb etc. would evaporate in the high temperature zone of the furnace, condense and enrich especially on the small ash particles in the down stream and could be exhausted into atmosphere from the system, because of the relatively low collection efficiency for the very small submicron particles in an electrostatic precipitator. So, it is very important for the development of a system of low pollutant emissions to make clear the physicochemical characteristics of ash particles and its correlations with the combustion conditions, coal properties and collection performance of electrostatic precipitator.

Recently, there have been reported the investigations on the ash formation processes and enrichment of trace elements on the particle surface. However, there are few reports which discuss the performance of electrostatic precipitator with considering

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physisochemical processes of particle formation in the furnace and its down-stream.

From the point of view above, this study is intended to investigate the physisochemical properties of various types of ash particles, especially enrichment behavior of sulfur, and their effects on the collection characteristics of ash particles in an electrostatic precipitator by use of an experimental system composed of one-dimensional pulverized coal combustion furnace, cyclone, bag-filter and cylindrical electrostatic precipitator. This type of combustion furnace has a large advantage of giving the pure effects of coal properties and experimental conditions on the ash particle formation.

2. Experimental system and procedure

A schematic of the experimental system used in this study is shown in Fig. 1. The method of one-dimensional combustion was adopted to eliminate the effect of turbulent mixing on the properties of exhaust gas and particles, and so the pure effects of coal properties on the particle formation could be derived in this system. The mixture of coal dust and air enters into the combustion furnace through a radiation shielding grid and burns one-dimensionally. Experiments were performed under the condition of excess air ratio of 1.2 and maximum temperature of 1500°C, which corresponds to that of practical boilers, and the coal feeding rate was about 3 kg/h. As a fuel, eight different kinds of pulverized coals shown in Table 1 were used.

Almost of the exhaust gas flows through a cyclone and bag-filter and is exhausted into atmosphere, and a part of it (about 15% by volume) is branched after the ash reservoir and introduced into the cylindrical electrostatic precipitator. The temperatures of cyclone and bag-filter are maintained to be 450°C and 100°C, respectively. The electrostatic precipitator consists of a discharge electrode wire of 0.4 mm in diameter set at the center line of the cylinder and a cylindrical collecting electrode which is 70 mm in inner diameter and 630 mm in length piled of nine cylindrical stages. At the exit of the electrostatic precipitator, a back-up filter is set to collect the sub-micron fine particles passed through the stages of electrostatic precipitator. The whole part of the electrostatic precipitator is heated up to be 150°C. Combustion products flows from the bottom to top and the gas flow velocity was

| Coal | Proximate analysis (%) | Ultimate analysis (% dry) | HHV (MJ/kg) |
|------|------------------------|--------------------------|-------------|
|      | M | Ash | VM | FC | C | H | O | N | S |                |
| Japan | A | 5.5 | 14.9 | 42.0 | 37.6 | 65.4 | 5.47 | 12.5 | 0.79 | 0.21 | 26.5 |
|       | B | 3.2 | 19.6 | 40.0 | 37.2 | 62.8 | 5.16 | 10.6 | 0.92 | 0.30 | 26.1 |
|       | C | 1.4 | 17.7 | 38.5 | 42.4 | 67.0 | 5.00 | 7.4 | 0.66 | 2.70 | 28.9 |
| China | D | 5.8 | 7.8 | 27.8 | 58.6 | 77.3 | 3.98 | 9.0 | 0.79 | 0.70 | 29.7 |
| Australia | E | 4.4 | 10.1 | 26.1 | 59.4 | 73.6 | 4.08 | 9.8 | 1.60 | 0.35 | 28.8 |
|       | F | 3.0 | 10.8 | 32.5 | 53.7 | 73.3 | 4.53 | 9.0 | 1.59 | 0.51 | 29.5 |
|       | G | 3.7 | 8.8 | 34.3 | 53.2 | 75.2 | 5.04 | 8.8 | 1.48 | 0.38 | 30.5 |
| South Africa | H | 2.4 | 14.8 | 25.5 | 57.3 | 70.5 | 3.64 | 8.3 | 1.64 | 0.99 | 28.1 |

M: Moisture, VM: Volatile matter, FC: Mixed carbon, HHV: High heat value
set to be 1.0 m/s in this experiment. After the experiment, ash particles collected in the electrostatic precipitator were recovered and the weight fraction of them in each stage was measured. Particles collected in the cyclone, bag-filter and each stage of the electrostatic precipitator were quantitatively analyzed by X-ray microanalyzer (XMA) and physicochemical features of them were systematically investigated. In the quantitative analyses by XMA, NBS 1633a was used as a standard sample, and elimination of background, separation of the overlap of peaks and many other important corrections (ZAF correction) were performed by use of a microcomputer in order to obtain the individual characteristics of X-ray intensity. Weight fraction of unburned carbon in the collected ashes were also measured by the method defined by JIS standard.

3. Characteristics of ash particles

The physical and chemical analyses of ash particles could give us a lot of informations about their formation processes. In this chapter, the characteristics of ash particles relating to the combustion and ash formation processes are discussed, and the attention is especially focussed on the surface concentrations of elements on the ash particles of various shapes and structures.

Among the ash particles collected in the down-stream devices, two typical shapes of spherical and non-spherical particles are observed. The surfaces of spherical particles almost look like smooth, while non-spherical particles are classified into two types of surface structures of smooth and porous. The typical SEM photographs of these types of particles obtained by use of coal A are shown in Fig. 2. The Photo (a) shows a spherical smooth particle sampled in the cyclone, (b) a non-spherical porous particle in the same stage and (c) both spherical and non-spherical particles with smooth surface in the bag-filter. The Photo (d) was taken after oxidizing the bag-filter ash particles in an electric furnace and powdery materials observed on the particles in Photo (c) have disappeared. So, these powdery materials on the bag-filter ash particles could be considered as soot substances. Every photo has the cross marks and numbers. The marks represent the analyzed points by XMA and, for examples, energy dispersive XMA spectra obtained for the points 1, 3 and 5 are shown in Figs. 3 (a), (b) and (c), which give the informations of the surface concentrations of elements indicated.

By the comparisons of the results obtained for the points 1 – 6, the following well-defined characteristics of selective existence of elements depending on the shapes and surface structures are noticed. The result at point 1 on the smooth spherical particle in the spectrum (a) shows that the most part of surface elements are occupied by Si and Al, and Ca, Fe, Mg and P follow those two elements in order. The points 2, 4 and 6 also give almost the same tendencies. The elements and their concentrations mentioned above could be considered to be typical for the smooth spherical particles. On the contrary, at the point 3 on the porous surface of non-spherical particles as shown in the spectrum (b), the concentrations of the elements other than Si and Al, especially sulfur, become higher than those at the point 1. Further, at the point 5 on the smooth surface of
non-spherical particles, Si predominates over other elements as shown in the spectrum (c).

These elements analyzed above are considered to exist on the particle surface in the form of oxides, such as SiO₂, Al₂O₃, Fe₂O₃, CaO and so on. Ca(OH)₂ and CaSO₄ formed under the conditions with H₂O and SO₂ would largely contribute to the electrical conductivity and dielectric constant of particles, which affect the collection efficiency of ash particles in an electrostatic precipitator. From the results mentioned above, we can conclude that the materials which could have much influence on the performances of electrostatic precipitator might be not all over but locally condensed on the particles depending on the shape and surface structure of ash particles.

4. Enrichment of sulfur on ash particles

Various important elements on the surface of ash particles were quantitatively analyzed by XMA. Among them the authors are especially interested in the concentration of sulfur, which largely affects the performance of electrostatic precipitator. Though sodium is considered to be important from the same standpoint of view, the discussion on it has been eliminated in this study because of the uncertainty of its quantitative data.

Here, the enrichment factor is introduced defined as

\[ EF_x = \frac{(C_x/C_{ref})_{ash}}{(C_x/C_{ref})_{coal}} \]  \hspace{1cm} (1)

where, \( C_x \) denotes the concentration of element \( x \), and \( C_{ref} \) is that of the reference element in the sample. Si was selected as the reference element because of the stability of its concentration which is not affected by diameter and temperature history of ash particles.

Enrichment factors of sulfur on the various types of ash particles obtained for coal A are shown in Fig. 4. There are little effects of diameter and surface structure on the enrichment factor of sulfur for the cyclone ash. On the contrary, for the bag-filter ash, very large enrichment factors up to 5 appear on the porous surface of non-spherical particles, and enrichment factors for spherical particles increase with the decreasing diameter.

Figure 5 shows the effect of particle size on the enrichment of sulfur on the spherical parti-
Ash particles with soot on it and those without. Figs. 5. (a) and (b) are the results for coal A and H, respectively. In both cases of (a) and (b), it is clearly shown that enrichment has occurred selectively on the small particles of bag-filter ash, while little effect of particle size appears for cyclone ash. The differences between the two enrichment behaviors for cyclone and bag-filter ashes could be explained by that the condensation and enrichment process of sulfur must have proceeded through the path from cyclone (450°C) to bag-filter (100°C) and that, in this process, the effect of diffusion of gas-phase sulfur compounds to the particle surface is larger for the smaller particles. Sulfur condensed on the particle surface is considered to exist mainly as the form of H$_2$SO$_4$ formed in the presence of H$_2$O and SO$_3$.

The effect of the enrichment of sulfur on the small particles of bag-filter ash is stronger for high volatile coal A than for low volatile coal H as distinctly shown in Fig. 5. The reason of this result could be due to that the soot particulates formed and attached on the ash particle surface must have a strong effect on the condensation process of sulfur on the particles, because less soot materials were observed on the bag-filter ash of coal H than on that of coal A.

In order to confirm the validity of this assumption above, unburned carbon contents in the cyclone and bag-filter ashes were measured as a function of volatile matter content in the raw coal for eight kinds of coals shown in Table 1. The result obtained is shown in Fig. 6. Cyclone ashes have lower unburned carbon content for higher volatile coals because high volatile coal has higher combustibility. On the other hand, the bag-filter ashes have higher unburned carbon content for higher volatile coals. The discrepancy of the unburned carbon content clearly corresponds to the gas-phase induced soot formed from hydrocarbons in the evolved volatile matter. This means that much more soot would have attached on the ash particles of high volatile coal A than low volatile coal H. The importance of the effect of soot particulates on the enrichment of sulfur could be also proved by the additional experiments in an electric furnace as described in the next section.

Ash particles with soot on it and those with-
5. Collection characteristics of electrostatic precipitator

In this chapter, collection characteristics of ash particles in an electrostatic precipitator are investigated taking into consideration the physicochemical characteristics of ash particles, especially the enrichment behavior of sulfur described above. The ash particles obtained in the electrostatic precipitator were observed by SEM and analyzed by XMA, and the weight fraction of collected ash particles in each stage (from first to ninth) of electrostatic precipitator was measured.

For spherical particles, the drift velocity of a particle in an electrostatic precipitator is given as

$$v = \frac{e_0 E_0 E_p}{3 \mu} k_\alpha d_p$$  \hspace{1cm} (2)

and the collection efficiency comes to be higher for the larger drift velocity. Here, dielectric coefficient $k_\alpha$ is given as

$$k_\alpha = 3 \epsilon_s/(\epsilon_s + 2)$$  \hspace{1cm} (3)

and is a weak increasing function of $\epsilon_s$, which is larger for higher concentration of sulfur on the particle surface. From Eqs. (2) and (3), it can be seen that the drift velocity is larger and then the collection efficiency is higher for larger particles and larger enrichment of sulfur. For this reason, by the results of SEM observation, almost all relatively large non-spherical and porous particles were collected at the first stage of electrostatic precipitator and particles...
collected in the following stages (second to ninth) are mostly spherical, the diameter of which rapidly becomes smaller along the latter stages.

The dependence on the particle size of the enrichment factors of sulfur on the surface of spherical particles collected in the first and ninth stages and back-up filter are shown in Fig. 8 for low sulfur coal A and high sulfur coal C. The tendency of sulfur enrichment on the smaller particles appears in each ashes, though the enrichment behavior is very much different by the coal type. For low sulfur coal A, the enrichment process have finished before the entrance of electrostatic precipitator, and so the particles collected in the earlier stage have larger enrichment factors of sulfur. And, for the particles with the same enrichment factors of sulfur, the larger particles have collected in the earlier stages. These results agree very well with the description of Eq. (2) for the drift velocity.

On the contrary, for high sulfur coal C, there must still have been the enrichment processes even in the electrostatic precipitator and enrichment behavior is very much complicated. The small submicron particles have a low collection efficiency as indicated by Eq. (2) and so those particles have higher enrichment factors of sulfur for the particles collected in the latter stages. For the particles from 1 \( \mu m \) to 3 \( \mu m \), particles of the ninth stage have a larger enrichment than those of back-up filter and this result could be due to that sulfur enriched particles have been selectively collected in the earlier stages because of higher dielectric constant. The particles larger than 3 \( \mu m \) have low and almost constant enrichment factors.

For the ashes of coal C which have a very complicated but interesting behaviors of sulfur enrichment, the weight fractions of ashes collected in each stage of electrostatic precipitator are shown in Fig. 9 by a bar graph. The weight fraction of ashes collected at the back-
up filter was very small and almost all ashes have been collected in the collecting electrodes. From this result, we can see the well-known tendency that the weight fraction decreases exponentially from bottom to top.

The solid line in Fig. 9 is the calculated result with the assumption that every particles have the same chemical and electrical properties and that the drift velocity of Eq. (2) depends only on the particle size. As for the initial condition for the particle size distribution at the entrance of electrostatic precipitator, the measured data by the image analyzer were used. However, there exists a large discrepancy between the solid line and the measured results.

On the other hand, the dashed line in Fig. 9 was obtained by introducing the effect of sulfur enrichment process in the electrostatic precipitator as shown in Fig. 8(b). In this case, sulfur enrichment behaviors in the electrostatic precipitator which affect dielectric constant of particles and then drift velocity of them have been taken into consideration according to Eqs. (2) and (3). This result of dashed line agrees much better with the measured results than the solid line.

From the results above, we can emphasize that it is very important to consider the detailed enrichment processes of elements such as sulfur, which have a strong effect on the electrical properties of ash particles, in order to predict the correct collection performance of electrostatic precipitator in a pulverized coal combustion system.

6. Conclusions

The correlations between physicochemical properties of ash particles and collection characteristics of particles in an electrostatic precipitator have been investigated with focusing on the enrichment behavior of sulfur and considering the effect of coal properties and combustion conditions on it. The following important conclusions have been obtained.

1) Chemical properties of ash particles largely depend on their shapes and surface structures. Spherical particles mainly consist of Al and Si, non-spherical particles with smooth surface are exclusively made of Si, and on the surface of porous particles there exist rather high concentrations of volatile elements such as S and Cl.

2) The enrichment and recondensation processes of sulfur which have evaporated in the combustion process would proceed with decreasing temperature in the downstream and largely depend on the temperature history of the particles. The enrichment factors become higher for smaller particles, if they are spherical, and for the particles with porous surface.

3) The tendency of sulfur enrichment on the smaller spherical particles are much stronger for high volatile coals than for low volatile ones. This is due to the fact that gas-phase induced soot formed from volatile matter and attached on the ash particles largely enhances the enrichment of sulfur on them.

4) The enrichment behaviors of sulfur on the ash particles have a strong effect on their collection characteristics in an electrostatic precipitator. Especially for the case where the enrichment process still proceeds in an electrostatic precipitator, it is essential to consider their effects on the changes of electrical properties of particles in the electrostatic precipitator in order to grasp its detailed performances.

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Nomenclature

\[ C_x \] : mass fraction of element \( x \) 
\[ d_p \] : diameter of ash particle [m]
\[ E_o \] : electric field of particle charging [V/m]
\[ E_p \] : electric field near collecting electrode [V/m]
\[ EF \] : enrichment factor [-]
\[ k_o \] : dielectric coefficient defined by Eq. (3) [-]
\[ VM \] : volatile matter content in coal [%]
\[ v \] : drift velocity of ash particles [m/s]
\[ \varepsilon_0 \] : dielectric constant of free space [F/m]
\[ \varepsilon_r \] : relative dielectric constant of ash particles [-]
\[ \mu \] : viscosity of exhaust gas [kg/m·s]

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