The current situation and subject of the fuel gas desulfurization process

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Coal is an important energy source in East Asia. Acid rain caused by emissions of sulfur dioxide is an environmental issue on a global scale. Environmental protection is the most important subject here. This author investigated environmental regulations and the current situation of flue gas desulfurization (FGD) at coal-fired power plants. The author compared the advantages of main FGD processes: the limestone gypsum process, the sea water washing process, the catalyst SOx oxidation process (CASOX), the activated carbon heating desorption process, the lime-base spray dry process, the furnace limestone injection process and electron beam. In order to select an appropriate FGD process, it is necessary to compare both the technological and economical advantages. Consequently, since the limestone gypsum process has a performance of more than 95% removal of sulfur dioxide, it is suitable for a large capacity coal-fired power plant. The lime-base spray dry process and CASOX process are simple, so they are more suitable for a small capacity coal-fired power plant.

Key Words: flue gas desulfurization, clean technology, sulfur dioxide, process selection

1. Introduction

Coal is an important energy source for power generation in East Asia. Acid rain caused by emissions of sulfur dioxide from coal combustion is an environmental issue on a global scale. Especially, technology for environmental protection at coal-fired power plants is the most important subject here. In order to protect the environment, Japan and China have developed new technologies; however, China desires the domestic technology of flue gas desulfurization (FGD).

In Japan, various FGD processes have been developed and commercialized since the 1970s. As a result, sulfur dioxide concentration in the ambient air has decreased.

The author has investigated environmental regulations, environmental protection systems, and the current situation of FGD at coal-fired power plants. The author has also investigated the performance of various FGD processes such as the limestone gypsum process, the sea water washing process, the catalyst SOx oxidation (CASOX) process, the activated carbon heating desorption process, the lime-base spray dry process, and the furnace limestone injection process.

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Most Japanese electric power companies have adopted the limestone gypsum process. Since China covers a large area, every region has its own economic situation regarding coal quality, the utilization of desulfurization reagents, and environmental protection regulations. To select an FGD process, it is necessary to compare the technological and the economical advantages of each.

2. Air environmental regulations

In Japan, energy use accompanied by economic growth has increased since the 1960s. In addition, air pollution of sulfur dioxide became a social problem. For this reason, the Clean Air Act was enacted in 1967. The regulation of sulfur dioxide emissions for power plants is determined by Equation (1). In this equation, q is the regulation of sulfur dioxide emissions ($m^3_N \cdot h^{-1}$), K is the coefficient classified by area, and He is the effective stack height. On the other hand, the local government has imposed an environmental preservation agreement with stricter regulations for electric power companies than those of the Clean Air Act.

$$q = K \times 10^{-3} \times H_e^2$$ (1)

Table 1 shows national emission standards of sulfur dioxide and values agreed upon by the local government and the Tsuruga coal-fired power plant of Hokuriku Electric Power.
Table 1  Emission standards of Tsuruga coal-fired power plant

<table>
<thead>
<tr>
<th>Item</th>
<th>Unit</th>
<th>No.1</th>
<th>No. 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unit size</td>
<td>MW</td>
<td>500</td>
<td>700</td>
</tr>
<tr>
<td>Emission standard of SO₂</td>
<td>m₃/h</td>
<td>919</td>
<td>1,095</td>
</tr>
<tr>
<td>Agreement</td>
<td></td>
<td>128</td>
<td>109</td>
</tr>
<tr>
<td>(coefficient in Tsuruga area)</td>
<td></td>
<td>8.0</td>
<td>8.0</td>
</tr>
<tr>
<td>Stack height</td>
<td>m</td>
<td>200</td>
<td>200</td>
</tr>
<tr>
<td>Effective stack height</td>
<td>m</td>
<td>339</td>
<td>370</td>
</tr>
</tbody>
</table>

China also made environmental protection a fundamental policy. In the Clean Air Act of China, the companies which emit sulfur dioxide have to pay a burden charge according to the regulation. The collected burden charge is used for air pollution prevention. As a result, the use of coal with low sulfur and low ashes, as well as clean coal technology, is promoted.

The new emission standards for coal-fired power plants were released in 2000.3) The emission standards were decided in three steps according to the construction period. Table 2 shows that the emission standards for the coal-fired power plants built after 2002 were the strictest standards. The emission standards before 2002 are somewhat less restrictive. This law will be enforced in 2010.

Table 2  Emission standards in China

<table>
<thead>
<tr>
<th>Item</th>
<th>Emission standard</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Per gas (mg/m³N)</td>
</tr>
<tr>
<td></td>
<td></td>
</tr>
<tr>
<td>Particulates</td>
<td>50</td>
</tr>
<tr>
<td>Inside of an acid rain regulation area</td>
<td>400</td>
</tr>
<tr>
<td>Outside of an acid rain regulation area</td>
<td>400</td>
</tr>
<tr>
<td>NOₓ</td>
<td>400~650</td>
</tr>
</tbody>
</table>

3. Environmental protection system in coal-fired power plants

The electric power companies actively promote policies regarding fuel, equipment, and operating procedures. Emissions of air pollutants such as particulates, sulfur dioxide, and nitrogen oxide are reduced through an appropriate combination of these three elements. In Japan, continuous reduction of particulates, sulfur oxide, and nitrogen oxide emissions is being achieved through the use of high-quality fuels such as those containing less than 1% sulfur. Coal-fired power plants installed high-efficiency electrostatic precipitators which remove more than 99% of particulates and high-efficiency wet-type limestone gypsum processes which remove more than 90% of sulfur dioxides.

In China, coal with low sulfur, low ash, and high calories is selected. Advanced coal power plants install high-efficiency electrostatic precipitators.

4. Characteristics of FGD processes

Fig. 1 shows the track record of FGD processes. Although FGD can be performed by various processes, the advantages and disadvantages, in general, are as follows. The wet type limestone gypsum process has a high-efficiency of sulfur dioxide removal and an efficient use of alkaline desulfurization reagents. However, this process has a higher construction cost than the dry and semi-dry processes. It is difficult to achieve a high-efficiency of sulfur dioxide removal with the dry and semi-dry FGD processes because the inside of a desulfurization reagent does not react with sulfur dioxide.

- **Limestone gypsum process**

Fig. 2 shows a conceptual flow of the limestone gypsum process.3) A limestone gypsum process has a track record of use in many large scale coal-fired power plants. This process has a performance of more than a 95% sulfur dioxide removal rate and maintained the performance regardless of the kind of coal used. However, this process requires a large quantity of water and, at the same time, a wastewater treatment facility. In addition, a flue gas re-heating system is needed for protection of the flue gas duct and stack from corrosion. This process increases the installation area, construction costs, and operating costs compared with dry and semi-dry FGD processes.

This process is suitable for plants near limestone producing areas. It is necessary to investigate whether gypsum has an effective market value as cement or a building material.
4.2 Sea water washing process

Fig. 3 shows a conceptual flow diagram of the sea water washing process. The sea water added with limestone is used for absorption of sulfur dioxide from flue gas in a once-through absorber. The slight increase of dissolved gypsum in the discharged sea water is small enough to be ignored when compared with the natural background level.

Specifically, due to the absence of a crystallization process and solid waste products, this process is simple and inexpensive. When slaked lime is added to sea water, this process has a performance efficiency of more than 90%.

4.3 Lime-base spray dry process

Fig. 4 shows a conceptual flow diagram of the lime-base spray dry process. In this process, slaked lime slurry is injected into the flue gas duct. This process has an efficiency performance of about 70%. When a lot of quicklime is added into the flue gas duct, this process has an efficiency of more than 90%. This process is not suitable for a large-scale coal-fired power plant because the performance is lower than the wet-type FGD process and a lot of the by-product is not reusable.

4.4 Furnace limestone injection & water spray process

Fig. 5 shows a conceptual flow of the furnace limestone injection and water spray process. Limestone powder is injected directly into the furnace, in which the sulfur dioxide reacts. Water is injected into the flue gas after air heater. The unreacted lime increases its reactivity by the addition of the water, which increases the sulfur dioxide removal rate. This process has a performance efficiency of about 70%. This process has the high possibility of damaging parts of the flue gas facility, such as the electrostatic precipitator and the active reactor tower, because limestone powder is injected directly into the furnace. Its use is not widespread, and it is not suitable for a large-scale coal-fired power plant.

In this method, a small quantity of water is required. The by-product is a mixture of gypsum, limestone, and fly ash; hence, reuse is difficult. Because there is no re-heating of the flue gas and no wastewater treatment, this process is simple and cost effective.

4.5 CASOX process

Fig. 6 shows a conceptual flow of the CASOX process. The sulfur dioxide in the flue gas is
adsorbed by activated carbon, oxidized by oxygen, and converted to dilute sulfuric acid with water vapor. As the diluted sulfuric acid is continuously removed from the catalytic layer, this process can be used successively without a need for catalyst regeneration.

The CASOX process has the following advantages. First, the process can reduce energy consumption compared with the conventional FGD process because a large amount of equipment, especially large fans and circulation pumps, is not required. Second, this process can be easily operated and maintained because of its simple configuration. Third, this process can be operated without alkaline because the absorbent is water. However, it is necessary to consider the reuse of the sulfuric acid (about 20wt-%) as a by-product.

Fig. 6 Schematic process flow diagram of CASOX process

4.6 Activated carbon heating desorption process

Fig. 7 shows the activated carbon heating desorption process. Sulfur dioxide in the flue gas is absorbed by activated carbon in a desulfurization tower, adsorbed sulfur dioxide and adsorbed nitrogen are desorbed in a desorption tower. Concentrated sulfuric acid is made from separated concentrated sulfur dioxide gas.

Fig. 7 Schematic process flow diagram of activated carbon heating desorption process

This process is a dry type and has the following advantages. A small amount of water is used. It is unnecessary to reheat flue gas. There is no scale and corrosion in equipment. Concentrated sulfuric acid is saleable. However, this process is complicated and the operating cost is high.

4.7 Electron beam process

Fig. 8 shows the electron beam process. This process can efficiently remove sulfur dioxide and nitrogen oxides by ammonia injection and electron beam irradiation. Sulfur dioxide and nitrogen oxides are oxidized by active species such as O, OH, and HO₂. They are neutralized in the gas phase by an ammonia reagent to form ammonium salts. This process consists of three principal steps: gas cooling of flue gas, followed by ammonia injection and electron beam irradiation, followed by by-product collection and reprocessing to an agglomerated form.

The by-product is saleable as manure with ammonium sulfate and ammonium nitrate. This process has a performance efficiency of more than 90%. Construction costs are high because of the electron beam. Moreover, operating cost is high due to the use of a lot of electric power and ammonia.

Fig. 8 Schematic process flow diagram of electron beam process

5. Conclusion

Table 3 shows the outline of the evaluation of several FGD processes. In China, the new emission standards for coal-fired power plants were released in 2000, and the enforcement is scheduled to begin in 2010. Treatments for flue gas emissions from coal-fired power plants, such as the FGD processes, are needed for companies.

The limestone gypsum process is suitable for a large-scale coal-fired power plant for the following reasons. First, a large-scale operation is possible. Second, limestone is abundant in Japan. Third, the gypsum by-product is saleable. This process consumes about 1% of the electric power generated, so it has been required to reduce energy consumption. Hokuriku Electric Power Company and Chiyoda Corporation jointly established new processes, such as
CASOX, to lower cost and decrease energy consumption.

The limestone gypsum FGD process with a removal efficiency of more than 95% is suitable for a large-scale coal-fired power plant, e.g., a plant with a capacity of more than 200 MW, where the sulfur content in the coal exceeds 2%. The lime-based spray dry FGD process with a removal efficiency of more than 75% is suitable for a small-scale coal-fired power plant, e.g., a plant with a capacity of less than 200 MW, where the sulfur content in the coal is under 2%.

The author has the following subjects. First, electric power companies do not have funds because adding to an electric bill is difficult. Second, the establishment of domestic technology of FGD processes is required. Third, it is necessary to establish a manufacturing method for alkaline desulfurization reagents. Fourth, it is necessary to reuse by-products. In Japan, a lot of gypsum caused problems in the early stages of the introduction of the limestone gypsum process.

References


Table 3 Characteristics of FGD processes

<table>
<thead>
<tr>
<th>Process</th>
<th>Scrubbing Medium</th>
<th>Removal Efficiency</th>
<th>Alkaline reagent</th>
<th>By-Product Name</th>
<th>Commercial Type</th>
<th>Waste water treatment</th>
<th>Amount of water</th>
</tr>
</thead>
<tbody>
<tr>
<td>Limestone gypsum</td>
<td>Wet</td>
<td>&gt;95</td>
<td>Limestone</td>
<td>Gypsum</td>
<td>Saleable</td>
<td>Needed</td>
<td>Large</td>
</tr>
<tr>
<td>Sea water washing</td>
<td>Wet</td>
<td>90</td>
<td>Sea water, Slaked lime</td>
<td>Non</td>
<td>Discharg</td>
<td>Needed</td>
<td>Large (Sea water)</td>
</tr>
<tr>
<td>Lime-base spray dry</td>
<td>Semi-dry</td>
<td>70~90</td>
<td>Quick lime</td>
<td>Limestone - Gypsum Mixture</td>
<td>Disposal</td>
<td>Non</td>
<td>Middle</td>
</tr>
<tr>
<td>Furnace limestone injection &amp; Water spray</td>
<td>Semi-dry</td>
<td>70</td>
<td>Limestone</td>
<td>Limestone - Gypsum Mixture</td>
<td>Disposal</td>
<td>Non</td>
<td>Middle</td>
</tr>
<tr>
<td>CASOX</td>
<td>Semi-dry</td>
<td>&gt;95</td>
<td>Water</td>
<td>Sulfuric acid</td>
<td>Saleable</td>
<td>Non</td>
<td>Large</td>
</tr>
<tr>
<td>Activated carbon heating desorption</td>
<td>Dry</td>
<td>&gt;95</td>
<td>Active carbon</td>
<td>Sulfuric acid</td>
<td>Saleable</td>
<td>Needed</td>
<td>Small</td>
</tr>
<tr>
<td>Electron Beam</td>
<td>Dry</td>
<td>&gt;90</td>
<td>Ammonia</td>
<td>Ammonium sulfate</td>
<td>Saleable</td>
<td>Non</td>
<td>Small</td>
</tr>
</tbody>
</table>