The whole process test and analysis of flue gas SO$_3$ in coal-fired power plant under variable load conditions

Zhigang Hong$^{1,2,3}$, Quankai Huang$^1$, Wei Chen$^1$ and Yongsheng Liu$^1$

$^1$Solar Institute of Shanghai University of Electric Power, Shanghai 200090, China;
$^2$Huadian Electric Power Research Institute Co., Ltd., Hangzhou 310030, China

$^3$Email: 1753990151@qq.com

Abstract. Under the current situation of ultra-low emission, it is of great significance to study the formation and removal of SO$_3$ in various equipments of coal-fired power plants. At present, most researches on SO$_3$ in coal-fired power plant focus on the influence of single equipment on flue gas SO$_3$ under a certain load condition. In this research, a 300MW generating unit is tested under variable load conditions to explore the variation law of SO$_3$ in various equipments under variable load conditions. The results show that SO$_3$ is mainly generated in the boiler furnace and SCR denitification device, and the total removal rate of SO$_3$ is 84.6%. However, the removal efficiency of SO$_3$ in electrostatic precipitators and the wet desulfurization systems shows great differences. Under full load condition, the removal rate of SO$_3$ is 46.8% - 69.8%. When the load changes, the SO$_3$ removal rate changes with the change of load, and the ESP is affected by the load most. The SO$_3$ content in the ash increases from 11.267mg/g under 50% load to 14.763mg/g under 100% load, and such changes are mainly determined by flue gas temperature and flue gas flow.

1. Introduction

The treatment of pollutants in coal-fired power plants has always attracted attentions. With the implementation of ultra-low emissions, the industry has gradually turned its focus to unconventional pollutants such as mercury, ammonia, SO$_3$ and condensable particles. Shi, Walsh P M et al believe that among the unconventional pollutants SO$_3$ has a huge impact on the environment and is the main cause of acid rain [1-2]. Menasha J, et al think in Selective catalytic reduction (SCR) denitrification system of coal-fired power plant, there is a certain amount of ammonia escaping, which will react with SO$_3$ in flue gas and generate ammonium bisulfate. The generated ammonium bisulfate and the particles in the flue gas continuously coagulate on the surface of the heat exchange element, resulting in the blockage of the air preheater [3]. Besides, the fly ash containing hydro sulphate will also cause SCR catalyst blockage; when the fly ash containing hydro sulphate passes through the bag filter, it will block the filter hole of the equipment, thus reducing the life span of the filter bag [4]. In view of the damage of SO$_3$ in coal-fired power plants to generating units and the pollution of the external environment, it is of great significance to study the specific distribution of SO$_3$ in coal-fired power plants and factors influencing its distribution.

Researchers have conducted extensive researches on the generation of SO$_3$ in the boiler and the effect of a single flue gas treatment equipment on SO$_3$. When the load of the unit changes, the conversion rate of SO$_3$ in the boiler furnace will change. At the same time, when the flow and flue gas temperature of the unit change, the conversion rate of SO$_2$/SO$_3$ in the catalyst in SCR will also change,
and the catalyst will affect the escape rate and conductivity of ammonia. The concentration of SO$_3$, ammonia escape rate and flue gas flow rate are closely related to the unit load. However, there are few kinds of research on the change of SO$_3$ in the whole process of coal-fired power plant. With a 300MW subcritical unit as a subject, this research studies the actual change of SO$_3$ in different removal equipments under the condition of variable load, and analyzes the change of SO$_3$ content of fly ash in the coal-fired power unit. It aims to explore the relationship between SO$_3$ and load of each test point in coal-fired power plant, and provide some reference for the subsequent researches and engineering practice of SO$_3$ emission reduction.

2. Material and methods

2.1. Plant description and feed fuels

The measured capacity of coal-fired unit is 300 MW, and the boiler is a SG-1025/17.5-M869 subcritical drum boiler. The boiler adopts tangential combustion from four corners. The coal type is medium slagging bituminous coal; direct-fired pulverizing system. The Denitrification system adopts high ash SCR arrangement, the reducing agent is urea, and the electrostatic precipitator, wet desulfurization system and wet electrostatic precipitator pollutant control equipment are used. The coal quality remained stable and the results of coal analysis during the test are shown in Table 1.

| Table 1. Coal proximate analysis. |
|----------------------------------|
|  | elemental analysis w/% | Industrial analysis w/% | calorific value/(MJ/kg) |
|  | M$_t$ | M$_{ad}$ | V$_{daf}$ | A$_d$ | C$_{ar}$ | H$_{ar}$ | N$_{ar}$ | O$_{ar}$ | S$_t$ | Q$_{netar}$ |
|  | 11.6 | 2.76 | 35.9 | 22.23 | 59.62 | 3.68 | 1.07 | 7.82 | 1.12 | 22.34 |

2.2. SO$_3$ detection method

2.2.1. The detection method of SO$_3$ in flue gas. At present, as shown in the literature [5], the sampling methods for SO$_3$ in coal-fired flue gas generally recognized and widely used in the field of environmental protection mainly include controlled condensation method and isopropanol absorption method. Through the investigation of related literature, the control condensation method is selected for sampling and testing. The sampling principle of the controlled condensation method is shown in Figure 1. Firstly, the flue gas is drawn out of the flue through a sampling gun at the same speed; after sampling, the sample gas flows through the filter device, and then the SO$_3$ in the flue gas is controlled and condensed, and finally the sampling device is washed and collected by deionized water. The SO$_3$ collected by condensation method will be converted into SO$_4^{2-}$ after process, and the SO$_3$ content in the corresponding flue gas can be obtained by measuring SO$_4^{2-}$.

Li Bin, Yang et al considered the sampling time, flow rate, heat tracing temperature, condenser pipe diameter, number of turns of SO$_3$ will affect the actual measurement [6]. In this study, the field test is carried out with a combination of the twisted tube (the coil diameter is 90 mm, inner diameter of tube is 5.5 mm, the pitch is 15 mm, number of coils is more than 23) and filter membrane filtration device; the sampling flow is controlled at about 20 L/min, and the sampling time is 45 min.

2.2.2. Detection of SO$_3$ in ash. The detection of SO$_3$ in ash is conducted mainly through the sampling, dissolution and measurement of ash. The ash sampling process is shown in Figure 2. During the sampling process, it is necessary to ensure that the samples are taken under the same working condition, and the uncontaminated sampling pipe is selected. After the sampling, the unified collection is carried out. The collected fly ash is completely dissolved in deionized water, and then analysed by gravimetric titration.

The content (%) of sulphur trioxides in ash by weight method is calculated according to the following equation (1):
In the formula, $m \rightarrow$ analysis ash sample weight; $m_1 \rightarrow$ blank test barium sulphate precipitation weight; $m_2 \rightarrow$ test barium sulphate precipitation weight;

$$W(\text{SO}_3) = \frac{0.343 \times (m_2 - m_1)}{m} \times \frac{250}{50} \times 100\%$$

(1)

Figure 1. SO$_3$ sampling flow chart as the graphic.

Figure 2. Schematic Diagram of Ash Sampling Process.

2.3. Selection of measuring points

Each symbol in the flowchart represents the corresponding equipment name: **SCR**: Selective Catalytic Reduction System; **APH**: Air Preheater; **ESP**: Electrostatic Precipitator

**WFGD**: Wet Flue Gas Desulfurization; **WESP**: Wet Electrostatic Precipitator

During the operation of the coal-fired flue gas in the coal-fired power plant, SO$_3$ sampling in the flue gas is shown in Figure 3, which is tested at the inlet and outlet of each flue gas treatment equipment. The sampling points of this sample are mainly coal economizer ash hopper, SCR outlet, air preheater outlet and dust collector ash hopper. Firstly, In the literature [7], the impact ash collection method can be used for collection. The sampling process is to insert the sampling gun vertically into the flue, and the sampling time is 20-30 min. When the sampling is finished, cool it to room temperature, and then seal and transfer it. For the air preheater outlet sampling, the boiler body sampling device is used, and the fly ash enters the sampling bottle through the cyclone separator. The sampling points of economizer and dust remover are relatively simple, for the sampling is conducted only at the corresponding ash hopper. To study the generation and control of SO$_3$ after the transformation of ultra-low emission, this test is carried out under the condition of variable load, which are 50%, 80% and 100% full load respectively.

2.4. Calculation of SO$_3$ generation in furnace flue gas

Zhang considered the mass concentration of SO$_3$ produced in the furnace of the generator set can be estimated according to formula 2 [8].
\[ CSO_3 = \frac{80}{22.4} \times 0.007 \times K_{SO_3} \times \frac{Sc, ar}{V_{dy}} \times 10^6 \times 100\% \]  

(2)

\( S_{c, ar} \) is the mass fraction of combustible sulphur in coal, \%; \( S_{c, ar} = 0.95St, ar \);
\( S_{t, ar} \) is the total sulphur content of coal, \%; \( V_{dy} \) is the mass volume of dry flue gas, m\(^3\)/kg;
\( C_{SO_3} \) is the \( SO_3 \) mass concentration in flue gas, mg/m\(^3\) (standard state).
\( K_{SO_3} \) is the conversion rate of \( SO_2/SO_3 \) during combustion (the sulphur content in coal is 0.5%-2.0%);
\( K_S \) is the correction coefficient (generally 0.90-0.95) for the conversion of combustible sulphur to Incombustible sulphur during combustion;

3. Results and discussion

3.1. Full flow analysis of \( SO_3 \) in flue gas under full load

3.1.1. Effect of boiler on \( SO_3 \) under full load. As shown in Figure 4, the total \( SO_3 \) removal rate of coal-fired unit is 84.6%. When the coal is burned in the boiler furnace, Srivastava RK et al. considered almost all combustible sulphur in the coal can be oxidized and burned to generate gaseous \( SO_2 \), in which about 0.5% – 2.0% of gaseous \( SO_2 \) will further oxidize to generate \( SO_3 \) [9]. At the same time, \( SO_2 \) produced in the process of partial combustion will further generate \( SO_3 \) under the catalysis of fly ash and metal oxides on the pipe wall. However, in the literature [10], the catalytic conversion of \( SO_2 \) in the furnace is closely related to the temperature, which is the highest at about 700 °C. According to the literature [11], due to the uneven distribution of \( SO_2 \) concentration in the boiler, the temperature and residence time distribution of \( SO_2 \) in various parts of the boiler have a close impact on the conversion rate of \( SO_3 \). The maximum relative deviation between the measured value of \( SO_3 \) collected from the furnace outlet, and the calculated value of formula 2 is only ± 2.46%. It can be seen that the selected \( SO_3 \) sampling system has high accuracy.

3.1.2. Effect of SCR system on \( SO_3 \) under full load. In coal-fired power plants, the generation of \( SO_3 \) in SCR system cannot be ignored. SCR is mainly used for the removal of nitrogen oxides in coal-fired power plants. According to the literature [12], Its working principle is to generate nitrogen by the oxidation-reduction reaction between nitrogen oxides and ammonia using the catalyst. However, the \( V_2O_5 \) in the catalyst can oxidize \( SO_2 \) to \( SO_3 \), which can increase the density of \( SO_3 \) in the flue gas. Some researchers think that the conversion rate of \( SO_3 \) in the SCR system is about 0.5% - 1.5% [13]. As shown in Figure 4 and Figure 5, the quantity of \( SO_3 \) in flue gas changes significantly when flowing through SCR, and the content of \( SO_3 \) increases from 30.13mg/m\(^3\) to 57.36mg/m\(^3\) under full load. It can be attributed to the oxidation of \( SO_2 \) in flue gas under the action of high temperature and catalyst in SCR.

3.1.3. Removal of \( SO_3 \) by ESP under full load. When the flue gas flows through ESP, the \( SO_3 \) removal rate reaches 46.8% at full load. This is mainly due to the following impact. With the existence of pits.

![Figure 4. Mass concentration of \( SO_3 \) at inlet and outlet of each equipment under full load.](image1)

![Figure 5. The removal rate of flue gas \( SO_3 \) in each equipment under full load.](image2)
and cavities on the surface of fly ash and the effect of electrostatic force and other forces on the pore wall of pores, SO$_3$ and moisture in flue gas form sub-micron sulfuric acid droplets through homogeneous nucleation and heterogeneous nucleation. Sulfuric acid droplets are easy to be adsorbed in the fly ash surface and thus condensed, and fine particles will polarize and be condensed when passing through the electric field area. Finally, large particles are formed by coagulation. Under the action of the electric field, SO$_3$ in flue gas is removed with the removal of fly ash. Li et al considered the removal efficiency of SO$_3$ in ESP mainly depends on the concentration of fly ash, specific surface area and flue gas temperature [14]. Between SCR outlet and ESP inlet, due to the decrease of temperature in the pipeline, the quantity of SO$_3$ in flue gas decreases, which leads to the condensation of SO$_3$ in the pipeline and the condensation of fly ash in the pipeline. Therefore, the low-temperature dust collector can effectively reduce the SO$_3$ in the power plant.

3.1.4. Removal of SO$_3$ by WFGD under full load. In the literature [15], the removal efficiency of SO$_3$ from flue gas by wet desulfurization device is generally 20% - 50%. In this test, the removal rate of SO$_3$ is 48.6%, and thus the removal efficiency is not high. The main reason is that the flue gas temperature decreases rapidly after entering the wet desulfurization system, but the flue gas SO$_3$ is transformed into submicron sulfuric acid aerosol particles in the process of rapid cooling, and the lower dust concentration in the desulfurization tower is not enough to absorb sulfuric acid aerosol particles.

3.1.5. SO$_3$ removal by WESP under full load. The removal efficiency of WESP for SO$_3$ in flue gas is the highest, which can reach 69.8%. According to relevant researches at home and abroad, the removal efficiency of WESP for SO$_3$ in flue gas generally ranges from 65% to 95% [16]. This is mainly because of the high content of water vapor in WESP, under the action of discharge electrode tip, the water mist will break and refine, resulting in a large number of charged droplets in the electric field, which increases the probability of submicron particles colliding with electricity; under the action of the electric field, the submicron particles are more likely to move to the dust collector and be removed, while the flue gas SO$_3$ in WESP mainly exists in the form of submicron H$_2$SO$_4$ mist, so wet ESP has a good effect on SO$_3$ removal.

![Figure 6](image1.png) Figure 6. Changes of SO$_3$ concentration in flue gas in each equipment under variable load condition.

![Figure 7](image2.png) Figure 7. The removal rate of SO$_3$ in flue gas in various equipment under variable load condition.

3.2. Removal of SO$_3$ from flue gas under different loads

As shown in Figure 6, the SO$_3$ content of flue gas in SCR increases with the increase of load, which is mainly related to coal consumption and the operating temperature of the boiler. The SO$_3$ conversion rate in the furnace is generally lower than 1.5%. The main component of catalyst in the SCR system is V$_2$O$_5$ and other metal oxides. Zhu et al considered V$_2$O$_5$ plays a leading role in the removal of nitrogen oxides and has a strong catalytic oxidation capacity for SO$_2$ [17]. When the unit load is 50%, the content of SO$_3$ only increases from 9.18mg/m$^3$ at the inlet to 20.24mg/m$^3$ at the outlet. This is because the catalyst activity decreases when flue gas temperature declines, so the conversion rate of SO$_2$/SO$_3$ in flue gas is low. When the load is increased from 50% to 80%, the flue gas temperature becomes
higher, and the catalyst activity in SCR and the conversion of SO$_3$ increases, and the removal rate of SO$_3$ decreases from -106.3% to -132.2% of 80% load. When the unit load is 100%, the removal rate of SO$_3$ in flue gas is reduced to -93.5%. At this time, the temperature is no longer the limiting factor of SO$_3$ conversion rate, because when the load is increased, the amount of coal-fired flue gas becomes larger and the flow rate quicker, which will lead to shorter retention time of SO$_3$ in flue gas. Moreover, with the increase of load, the ammonia injection amount in SCR will increase, for NH$_3$ and SO$_2$ in SCR will compete under the action of catalyst. The increase of NH$_3$ content will inhibit the catalytic oxidation of SO$_2$ in SCR, so the catalytic efficiency of SO$_2$ will be reduced.

Figure 7 shows the content of flue gas SO$_3$ in ESP increases with the increase of load, the main reason is that the higher the load is, the greater the flue gas volume is; the removal rate of SO$_3$ changes significantly with the increase of load, and the removal rate is more than 40% under high load, while the ratio of full load to 80% load does not change significantly, and even decreases, and the root cause is the temperature and flow rate. The higher the flow rate, the shorter the residence time and the higher the temperature, the more difficult it is for SO$_3$ to be condensed and removed. In WFGD, the quantity of SO$_3$ changes less, and the removal rate of SO$_3$ increases with the increase of load. This is because in the wet desulfurization system, the temperature changes little under different loads, and the removal effect is mainly affected by the flue gas flow and flow rate. The quantity of SO$_3$ in WESP is less than 5mg/m$^3$, which shows that the effect of wet electrostatic precipitator on SO$_3$ removal under different loads is significant, and the difference of SO$_3$ removal rate under different loads is small.

3.3. Changes of SO$_3$ content in fly ash under different loads

Figure 8 shows the change of SO$_3$ mass fraction adsorbed in fly ash samples at each test site under full load. Because fly ash contains a large quantity of metal oxides such as calcium oxide and iron oxide, the surface of fly ash is usually alkaline and easy to absorb acid gases such as SO$_3$ and HCl. Due to the catalytic oxidation of SCR, the quantity of SO$_3$ in flue gas increases, so the quantity of SO$_3$ in fly ash increases sharply, directly from 5.162 mg/g to 12.935 mg/g. At the outlet of the air preheater, the mass fraction of SO$_3$ adsorbed in the ash is greatly reduced. This is due to the cooling effect of the air preheater on the flue gas, which causes SO$_3$ in the flue gas to condense to form sulfuric acid mist, or SO$_3$ reacts with ammonia in the flue gas to form ammonium bisulphate, which adheres to the surface of the air preheater, reducing the content of SO$_3$ in the flue gas, thus reducing the mass fraction of SO$_3$ adsorbed in the ash. The density of SO$_3$ is 14.736mg/g in the ash sample taken from the electrostatic precipitator, which is mainly because before entering the electrostatic precipitator the ash particle is large, the specific surface area of the fly ash is small, and the alkali matter dissolved in the water is less. The electrostatic precipitator has a good removal performance for the large dust particles in the flue gas. When the flue gas temperature gets lower, the adsorption capacity of acid gas on the surface of basic fly ash becomes stronger. The sulfuric acid mist formed by SO$_3$ is easy to combine with micron-level dust, resulting in the increase of the mass fraction of acid gas adsorbed in the ash, so the quantity of SO$_3$ in the ash after the dust collector is significantly increased in the test.

Figure 9 and Figure 10 shows that under variable load conditions, the mass fraction of SO$_3$ adsorbed in the ash is quite different. In the range of 4.9-15 mg/g, the mass fraction of SO$_3$ adsorbed in the ash is positively correlated with the mass concentration of flue gas. The greater the load is, the greater the flue gas quantity is, and the greater quantities of the SO$_3$ the fly ash absorbs. At the outlet of SCR system, the mass fraction of SO$_3$ adsorbed in ash is more than twice of that at the outlet of economizer. The reason lies in the catalytic oxidation of SO$_3$ by the SCR system and the increase of SO$_3$ content in flue gas, which makes the mass fraction of SO$_3$ adsorbed in ash increase. At the same time, due to the increase of load, temperature and flow rate, the content of SO$_3$ in ash increases. For air preheater, the change of SO$_3$ content is not obvious under the condition of variable load. The electrostatic precipitator affected by the load most. The quantities of SO$_3$ in the ash increases from 11.267mg/g of 50% load to 14.763mg/g of 100% load. The removal of SO$_3$ by ESP is still through the removal of dust particles in the flue gas. The quantities of SO$_3$ in the ash is closely related to the density of dust particles in the flue gas. The higher the load, the higher the content of SO$_3$ and dust.
particles, and the higher the adsorption capacity between the particles and SO\textsubscript{3}, thus improving the removal of SO\textsubscript{3} by ESP.

![Graph](image1.png)

**Figure 8.** Mass fraction of SO\textsubscript{3} in ash at each sampling point under full load condition.

![Graph](image2.png)

**Figure 9.** Mass fraction of SO\textsubscript{3} in ash at each sampling point under variable load condition.

![Graph](image3.png)

**Figure 10.** Gas temperature at ash sampling point under variable load condition.

4. Conclusions

The variation law of SO\textsubscript{3} in coal-fired power plant under variable load conditions is systematically studied by testing the whole process of SO\textsubscript{3} in a 300WM generating unit. By understanding the characteristics of SO\textsubscript{3} emissions, technical support can be provided for the next step of emission control for coal-fired power plants.

1) The experimental results show that the SO\textsubscript{2} concentration in the flue gas increases greatly under the catalytic oxidation of the catalyst in the SCR system, and the SO\textsubscript{3} content increases from 30.13mg/m\textsuperscript{3} to 57.36mg/m\textsuperscript{3}. Other pollutant removal equipment has a certain removal effect on SO\textsubscript{3} in flue gas. Under full load conditions, the removal rates of SO\textsubscript{3} in flue gas by ESP, WFGD and WESP are 46.8%, 48.6%, and 69.8%, respectively.

2) Under the conditions of 50%, 80% and 100% variable load, the density of SO\textsubscript{3} in flue gas is affected by flue gas flow, velocity and temperature. In SCR, the conversion rate of SO\textsubscript{2} in flue gas is the highest under 80% load. In ESP, the removal efficiency of SO\textsubscript{3} in flue gas is the best under 80% load. When the flow gas reaches WESP, its concentration becomes low, and there is no big difference of SO\textsubscript{3} removal under variable load condition.

3) Under the condition of full load, the content of SO\textsubscript{3} in fly ash can reach 4.9-15 mg/g. At the outlet of the air preheater, the quantity of adsorbed SO\textsubscript{3} in the ash drops sharply, mainly because part of the SO\textsubscript{3} reacts with the ammonia in the flue gas and adheres to the inner wall of the pipe. In the case of variable load, the quantity of SO\textsubscript{3} in ash is mainly affected by flue gas temperature and flue gas flow. The data shows that under normal circumstances, when the load increases, the quantity of adsorbed SO\textsubscript{3} in the ash will increase accordingly.
As the load changes, the quantity of other pollutants in coal-fired power plants will also change. Therefore, coal-fired power plants can adjust the load according to the actual emission demand, so as to achieve efficient and economic emission reduction effect.

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