Limestone-Gypsum Wet Flue Gas Desulfurization Wastewater Treatment

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Abstract. In this paper, chemical precipitation process is used for the treatment of the desulfurization wastewater generated by the waste gas treatment system of the limestone-gypsum flue gas desulfurization process of a thermal power plant. After one year of operation, the technology is reliable and stable. The removal rate of suspended matter, COD, fluoride, mercury and sulfate reached 99.5%, 98.3%, 87.5%, 99.8% and 85.6% respectively, and the corresponding effluent concentrations were 54 mg/L, 20 mg/L and 9.6 mg/L, 0.04 mg/L, 980 mg/L. Finally, the effluent quality of the system is better than the discharge requirements of “Discharge standard of wastewater from limestone-gypsum flue gas desulfurization system in fossil fuel power plants” (DL/T997-2006). In addition, the total electricity and chemical charges per ton of wastewater treatment were 6.50 yuan, which is lower than most of the engineering project under the same discharge requirements. Therefore, the process has certain engineering advantages.

1. Introduction
As the most popular desulfurization technology, limestone-based wet flue gas desulfurization (WFGD) has been widely used worldwide, which has about 85% of the market share in China [1-3]. However, although limestone-based WFGD benefits from its advantages of high stability, abundant raw materials and low operating cost, it is inevitable to produce a large amount of wastewater, which has serious pollution to the environment [4]. Therefore, it is necessary to research the treatment of limestone-gypsum WFGD wastewater.

It is an effective way to study the wastewater treatment by taking the actual engineering example as sample. In this paper, a thermal power plant with pulverized coal boiler of 3×240 t/h, 1×240 t/h and 1×460 t/h in China was chosen as a sample. To protect the living and production environment around the power plant, the supporting flue gas desulfurization device was built to treat the boiler flue gas to reach the discharge standard. The reliable process of limestone-gypsum WFGD with the configuration of five furnaces and three towers was adopted, and the absorption tower adopts countercurrent spray empty tower [5, 6]. The treatment object of this sample is the desulfurization wastewater generated by the waste residue treatment system of flue gas desulfurization process, with the wastewater volume of
about 600 m$^3$/d. The desulfurization wastewater has the following characteristics [7]: it is acidic, contains trace heavy metal ions, fluorine/sulfide exceeds the standard; the content of suspended solids (mainly ash particles, SiO$_2$ and iron/aluminum hydroxide) is high; COD is high, mainly caused by sulfite, Fe$^{2+}$ and other reducing substances and High Cl$^-$ (up to 18000-30000 mg/L) [8]; the acid substances and anions in wastewater mainly come from flue gas, while cations and heavy metal ions mainly come from limestone used for desulfurization [9, 10]. With the more and more strict requirements for the discharge of waste water in thermal power plants, it is necessary to strengthen the rational treatment of this kind of desulfurization wastewater with complex components and many kinds of pollutants.

2. Process and Methods

2.1. Wastewater quality

The selection of additive parameters and the optimization of treatment process are carried out according to the field investigation of the water quality and quantity of flue gas desulfurization wastewater in thermal power plant, and the practical experience of similar projects, literature reports and laboratory small/pilot test results, the "Chemical Precipitation Coagulation Method" is adopted to treat the desulfurization wastewater [11, 12]. The main water quality indexes and discharge requirements of the waste water of the thermal power plant are shown in Table 1.

| Index/water quality | pH | Suspended matter | COD | Fluoride | Total mercury | Total cadmium | Total arsenic | Total lead | Sulfate |
|---------------------|----|------------------|-----|----------|---------------|---------------|--------------|------------|---------|
| Effluent Quality Discharging standard DL/T997-2006 | 4.5~7.5 | 10740 | 1211 | 76.7 | 16.0 | 0.113 | 0.0046 | 0.491 | 6800 |
| Discharging standard DL/T997-2006 | 6-9 | 70 | 150 | 30 | 0.05 | 0.1 | 0.5 | 1.0 | 2000 |

2.2. Technological process

The desulfurization wastewater treatment process of this sample is shown in Figure 1.

As shown in Figure 1, the desulfurization wastewater flows into the regulating tank by itself, and is lifted to the sedimentation tank by the primary lift pump. After gravity sedimentation, the supernatant flows into the neutralization reaction tank. Lime milk solution and appropriate amount of HCl are added to adjust the pH value to 8.8-9.2, so that most heavy metal ions can form insoluble hydroxide precipitation in alkaline environment [13]. The waste water flows into the organic sulfur reaction tank by itself, adding organic sulfur (TMT15) to react with Hg$^{2+}$, Cd$^{2+}$ and other metal ions to form insoluble sulfide [14]. The effluent flows into the coagulation reaction tank, and the flocculant FeClSO$_4$ is added to make the fine particles in the wastewater agglomerate into large particles and settle down [15]; the effluent enters the flocculation reaction tank, and PAM is added to make the fine flocs become more settling flocs. The effluent flows to the inclined tube sedimentation tank, and the supernatant overflows to the transition tank after sedimentation. Aeration system is set in the transition tank to oxidize SO$_3^{2-}$ and other reducing substances in the wastewater through O$_2$. The effluent flows from the secondary lift pump to the sand filter tank and flows into the clean water area after filtration to meet the discharge standard. The sand filter tank is backwashed regularly, and the backwash water flows into the wastewater regulation sedimentation tank.

The sludge containing heavy metals in neutralization reaction tank and organic sulfur reaction tank shall be sent to qualified companies for treatment; ordinary sludge such as sedimentation tank and inclined tube sedimentation tank shall be transported to sludge tank by sludge pump, and then transported to plate and frame filter press by screw pump for dehydration and then transported out for disposal.
2.3. Design of process unit

2.3.1. Regulating tank. A new regulating tank with a design capacity of 30 m³/h was built. Intermittent drainage is carried out each day in the desulfurization section, and the regulating pool is used to regulate and store the uneven wastewater discharged by the desulfurization system, and balance the wastewater quality to ensure that the treatment system has a certain buffer capacity. At the same time, the sludge dewatering filtrate is also returned to the wastewater regulating tank and enters the wastewater treatment cycle together with the influent water.

Its size is 5.5 m×4.0 m×3.5 m, the effective water depth is 3.0 m, the effective volume is 66 m³, and the residence time is 2.2 h. Its material is steel concrete, and the inner wall is anticorrosive.

Supporting equipment: two sets of primary lifting pumps (one for use and one for standby with wide flow channel, the flow rate is 30 m³/h, the power is 5.5 kW, the material of overflow part is SS316L), two sets of negative pressure tanks (φ 0.4 m×1.0 m, the material is FRP), hyperbolic mixer (one set, the power is 2.2 kW, the diameter is 1 m, the speed is 70 r/min), one set of ultrasonic liquid level meter.

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**Figure 1.** Wastewater treatment process flow diagram
2.3.2. **Sedimentation tank.** There are two sets of sedimentation tank in total, running alternately, and the design scale is 30 m³/h. The content of suspended solids in wastewater is high, and the role of sedimentation tank is to precipitate suspended solids in wastewater. A mud scraper is set at the bottom of the sedimentation tank, and a decanter is set at the top. The supernatant flows into the neutralization reaction tank by gravity through the decanter. The size of the single tank is φ 5 m×8.5 m, the effective water depth is 8.0 m, the effective volume is 157 m³, and the residence time is 5.3 h. Its material is steel concrete, and the inner wall is anticorrosive.

Supporting equipment: two mud scrapers (PBX-5.0, the power is 1.1 kw); two water decanters (BSL-30, FRP, the power is 1.1 kw).

2.3.3. **Neutralization reaction tank.** There is one neutralization reaction tank, with a design capacity of 30 m³/h. The pH is adjusted into 8.8-9.2 by adding Ca(OH)₂ and HCl into the neutralization tank, so that most heavy metal ions would form insoluble hydroxide precipitation in alkaline environment [16]. At the same time, the fluoride can be removed as the Ca²⁺ and F⁻ in wastewater would react to form insoluble CaF₂; the arsenic content can be reduced as Ca²⁺ reacts with arsenic to form precipitated Ca₃(AsO₃)₂; the concentration of lime milk is 5%, and the dosage is about 4.5 g/L; the concentration of hydrochloric acid is 5%, and the dosage is about 8.5 ml/L. The size of neutralization reaction tank is 3.0 m×2.0 m×4.0 m, the residence time is 0.7 h, and the effective water depth is 3.5 m. Its material is carbon steel, and the inner and outer walls are anticorrosive.

Supporting equipment: one mechanical mixer (anticorrosive, the speed is 30 rpm, the agitator diameter is 600 mm, the power is 2.2 kW); one pH on-line monitor (including control system); one set of lime milk dosing system (one set of lime silo with size of φ 2.6 m×8.0 m and effective volume of 40 m³; two sets of lime milk reaction boxes with the size of φ 2.5 m×3.5 m and effective volume of 16 m³. There are two sets of lime milk storage tanks with the size of φ 2.5 m×3.5 m and effective volume of 16 m³; two Ca(OH)₂ lifting pumps, one for use and one for standby, with flow rate of 10 m³/h and power of 11 kw; two Ca(OH)₂ dosing pumps, one for use and one for standby, with flow rate of 1.8 m³/h and power of 2.2 kW; one air compressor with air displacement of 2.0 m³/min and power of 11 kw; one set of hydrochloric acid dosing system (two sets of dispensing devices, one for use and 1 for standby, with the size of φ 2.5 m×3.5 m, single set volume of 8 m³; two sets of mixers, power of 0.75 kW; two sets of hydrochloric acid dosing pumps, with flow rate of 0-1.2 m³/h, one for use and one for standby; one set of hydrochloric acid storage tank, with the size of φ 2.5 m×5.0 m and volume of 24 m³; one hydrochloric acid discharge pump, with flow rate of 7.2 m³/h and power of 2.2 kW).

2.3.4. **Organic sulfur reaction tank.** There is a total of one set of organic sulfur reaction tank, with design scale of 30 m³/h. Organic sulfur (TMT-15) is added to the reaction tank to make the heavy metal ions that are not easy to precipitate in the form of hydroxide form sulfide with smaller solubility product to precipitate [17]. The organic sulfur reaction tank has good treatment effect on wastewater containing cadmium, zinc, mercury, etc. as the solubility of heavy metal sulfide is small, the precipitation volume is small, the chemical stability is good, and it is not easy to dissolve back. The size is 3.0 m×2.0 m ×4.0 m, the residence time is 0.7 h, and the effective water depth is 3.5 m. The material is carbon steel, and the inner and outer walls are anticorrosive.

Supporting equipment: one mechanical mixer (anticorrosive, the rotating speed is 30 rpm, the agitator diameter is 600 mm and the power is 2.2 kW); one set of organic sulfide dosing system (one set of TMT 15 chemical storage tank with size of 2.0 m×0.5 m×0.5 m, volume of 500 L; one dispensing tank with size of 2.0 m×1.0 m and volume of 2 m³; one mixer with power of 0.75 kW; two dosing pumps with flow rate of 200 L/h and power 0.55 kW, one for use and one for standby).

2.3.5. **Coagulation reaction tank.** There is one set of coagulation reaction tank, with design scale of 30 m³/h. After the neutralization reaction and heavy metal precipitation reaction are completed, the coagulant FeCl₃ is added to the coagulation reaction tank to make the micro flocs in the wastewater form large flocs gently. Through flocculation, a large number of suspended particles which cannot be
directly precipitated in the wastewater can be destabilized and agglomerated to form large particles and removed [18]. The dosage of FeClSO₄ was 0.3 kg/m³, and the concentration was 10 %. The size of coagulation reaction tank is 3.0 m×2.0 m×4.0 m, the residence time is 0.7 h, and the effective water depth is 3.5 m. The material is carbon steel, and the inner and outer walls are anticorrosive.

Supporting equipment: one mechanical mixer (anticorrosive, the rotating speed is 60 rpm, the agitator diameter is 600 mm, the power is 4.0 kW); one FeClSO₄ dosing system (two sets of dispensing tanks with single size of 2.0 m×1.0 m×1.2 m, single set volume of 2 m³; two sets of mixers with power of 1.5 kW; two sets of dosing pumps, one for use and one for standby, the flow rate is 200 L/h, the power is 0.55 kW).

2.3.6. Flocculation reaction tank. There is one set of flocculation reaction tank with design capacity of 30 m³/h, PAM is put into the flocculation reaction tank as coagulant aid to further increase the floc and make it easier to settle [19]. The size is 3.0 m×2.0 m×4.0 m, the residence time is 0.7 h, and the effective water depth is 3.5 m. The material is carbon steel, and the inner and outer walls are anticorrosive.

Supporting equipment: one mechanical mixer (anticorrosive, the speed is 30 rpm, the impeller diameter is 600 mm, the power is 2.2 kW); one set of PAM dosing system (one set of PAM integrated dissolution and dosing device with pharmaceutical capacity of 2000 L/h; two PAM dosing pumps, one for use and one for standby, the flow rate is 480 L/h, the power is 0.75 kW).

2.3.7. Inclined tube sedimentation tank. There is one set of inclined tube sedimentation tank with design capacity of 30 m³/h, whose main function is to separate the sludge from water, settle the sludge into the sludge hopper at the bottom, lift it from the sludge pump to the sludge tank, and make the supernatant flows to the transition tank by itself. The size is 7.0 m×4.0 m×4.0 m, effective water depth is 3.4 m, surface load is 1.0 m³/(m²·h), solid load is 150 kg/(m²·d). The material is carbon steel, and the inner and outer walls are anticorrosive.

Supporting equipment: inclined pipe packing 28 m² (the diameter is 35 mm, the height is 800 mm, the material is ethylene propylene copolymer); one set of inclined pipe support; two sludge pumps (wide channel non-blocking sludge pump, the flow rate is 11 m³/h, the power is 1.1 kW).

2.3.8. Transition pool. There is a total of one transition pool with design capacity of 30 m³/h, and an aeration system is set inside to oxidize sulfite and other reducing substances in wastewater. The wastewater from the transition tank is lifted to the sand filter tank by the lift pump. The size is 4.5 m×4.0 m×4.0 m, the effective water depth is 3.2 m, the effective capacity is 57.6 m³, and the residence time is 2.0 h. The material is carbon steel, and the inner and outer walls are anticorrosive.

Supporting equipment: two roots fans with the flow rate of 1.65 m³/min, wind pressure of 0.04 MPa, power of 2.2 kW, one for use and one for standby; two waste water lifting pumps, one for use and one for standby (horizontal centrifugal pump with flow rate of 30 m³/h, power of 5.5 kw).

2.3.9. Sand filter tanks. There are two sand filter tanks, one for use and one for standby. The design scale of a single set is 30 m³/h with the size of φ 2.2 m×3.5 m, and the filtration speed of 8.0 m³/(m²·h). The filler is made of refined quartz sand with a filling height of 1400 mm, including the bearing layer with particle size of 3-5 mm, the intermediate layer with particle size of 1.2-2.0 mm, the upper layer with particle size of 0.5-1 mm; the backwashing cycle is once a day, and the backwashing time is set to 25 min; the backwashing strength is 8.5 L/(m²·s). The material is FRP, and the inner wall is anticorrosive.

Supporting equipment: one backwash water pump (horizontal centrifugal pump with flow of 240 m³/h, power of 30 kW).

2.3.10. Clean water tank. There is a total of one clean water tank with design capacity of 30 m³/h, which is used to store the filtered water from the sand filter tank, and the water from the clean water tank is used as the backwashing water for the sand filter tank. The size is 2.0 m×4.0 m×4.0 m, the effective
water depth is 3.5 m, and the residence time is 0.9 h. The material is carbon steel, and the inner and outer walls are anticorrosive.

2.3.11. Sludge dewatering system. A total of one sludge tank is set up. The sludge is lifted to the plate and frame filter press by screw pump for sludge dewatering and sludge cake transportation. The size of sludge tank is 3.0 m×3.0 m×4.0 m, and the effective volume is 42 m³. The material is carbon steel, and the inner and outer walls are anticorrosive.

Supporting equipment: two sets of high pressure diaphragm frame filter press (the filtering area is 700 m², the power is 14.95 kW); two screw pumps (the flow rate is 56 m³/h, the power is 30 kW); two press pump (the flow rate is 24 m³/h, the power is 30 kW); two sets of press water tank (the size is 2.0 m×1.5 m×1.5 m, the material is FRP); two sets of electric hoist (the lifting capacity is 5.0 t, the power is 5.5 kW); two sets of belt conveyor (the belt width is 500 mm, the conveying length is 10 m, the power is 4 kW).

3. Results and discussion

3.1. The effect of wastewater treatment

After commissioning, the desulfurization wastewater treatment system operates stably for 6 months with good treatment effect and stable effluent quality. The pollutant indexes can meet the design effluent quality requirements. The removal rates of suspended solids, COD, fluoride, mercury and sulfate can reach 99.5%, 98.3%, 87.5%, 99.8% and 85.6% respectively, and the corresponding effluent concentrations are 54 mg/L, 20 mg/L, 9.6 mg/L, 0.04 mg/L and 980 mg/L. The effluent is significantly better than the discharge standard of DL/t997-2006. The water quality of each main treatment unit is shown in Table 2.

Table 2. List of processing efficiency of the main structures

| Main processing unit                  | COD (mg/L) | suspended solids (mg/L) | fluoride (mg/L) | mercury (mg/L) | sulfate (mg/L) |
|---------------------------------------|------------|-------------------------|----------------|---------------|----------------|
| Sedimentation tank inlet              | 1211       | 10740                   | 76.7           | 16.0          | 6800           |
| Removal rate                          | 1130       | 6700                    | 74.5           | 15.6          | 6500           |
| Neutralization reaction tank:         | 6.7%       | 37.6%                   | 2.9%           | 2.5%          | 4.4%           |
| Removal rate                          | 920        | 5800                    | 19.9           | 14.4          | 5960           |
| Organic sulfur reaction tank:         | 18.6%      | 13.4%                   | 73.3%          | 7.7%          | 8.3%           |
| Removal rate                          | 680        | 4100                    | 14.5           | 1.2           | 5210           |
| Coagulation flocculation tank:        | 26.1%      | 29.3%                   | 27.1%          | 91.7%         | 12.6%          |
| Removal rate                          | 330        | 1760                    | 13.4           | 0.39          | 2350           |
| Inclined tube sedimentation tank:     | 51.5%      | 57.1%                   | 7.6%           | 67.5%         | 54.9%          |
| Removal rate                          | 105        | 260                     | 10.9           | 0.22          | 1270           |
| Sand filter tank:                     | 68.2%      | 85.2%                   | 18.7%          | 43.6%         | 46.0%          |
| Removal rate                          | 20         | 54                      | 9.6            | 0.04          | 980            |
| Total removal rate                    | 98.3%      | 99.5%                   | 87.5%          | 99.8%         | 85.6%          |

3.2. Technical and economic analysis

The total investment of the wastewater treatment system is 9.15 million yuan. The total installed capacity of the electrical equipment is 327.95 kW, and the operating power is about 291.8 kW. According to the
utilization efficiency of 60%, the power consumption is 5.836 kW·h /t per ton, the electricity charge is 0.7 yuan/kW·h, and the treatment power charge is 4.08 yuan/m³.

The unit prices of Ca(OH)₂, HCl, organic sulfur, FeClSO₄ and PAM are 300 yuan/t, 300 yuan/t, 7500 yuan/m³, 2000 yuan/t, 20000 yuan/t, respectively. The consumption of each ton of water treatment is 4.5 kg, 0.4 kg, 0.02 l, 0.12 yuan/m³, 0.15 yuan/m³, 0.60 yuan/m³, 0.20 yuan/m³, which is equivalent to 2.42 yuan/m³ under normal operation. As a result, the cost of water treatment is 6.50 yuan/m³.

4. Conclusion
For the limestone-based WFGD wastewater, the "Chemical Precipitation Coagulation Method" is adopted in this study, which greatly saves the investment, operation and maintenance costs compared with the "Three Effect Evaporation" and "Horizontal Film Spray/MVC Evaporation", and also reduces the difficulty of later operation.

The removal rates of suspended solids, COD, fluoride, mercury and sulfate can reach 99.5 %, 98.3 %, 87.5 %, 99.8 % and 85.6 % respectively, and the corresponding effluent concentrations are 54 mg/L, 20 mg/L, 9.6 mg/L, 0.04 mg/L and 980 mg/L. The effluent quality meets and is significantly better than the discharge requirements of “Discharge standard of wastewater from limestone-gypsum flue gas desulfurization system in fossil fuel power plants” (DL/T997-2006), which will reduce the pollution load brought by the effluent discharge to a greater extent. The total cost of electricity and medicament for the wastewater treatment is 6.50 yuan/m³, and its operation cost is lower than the treatment cost of 7.35 yuan/t under the same discharge requirements of Ref. [20]. Therefore, the process has certain engineering advantages.

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