Combined Removal Of NOx And SO2 From Flue Gas At Low Temperature

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Abstract. A method was proposed to remove NOx and SO2 in flue gas by using the sulfinyl functional group as a catalyst. Ozone is introduced into the flue gas to oxidize NO. Soluble NO2 and SO2 reacted with ammonia to form ammonium sulfate and ammonium nitrate, which were the raw material of the compound fertilizer. A small pilot is built in a container that can be easily transported to power plant and extracts the actual flue gas directly from the gas duct. In order to obtain the best the SO2 and NOx removal efficiency in this experiment, many parameters were changed. Such as flue gas flow, ozone / NOx ratio, liquid-gas ratio, flue gas temperature, catalyst type, catalyst concentration, solution pH value. Results indicated that SO2 was cleaned up quite efficiently and the removal efficiency was nearly 99% under all conditions. the best NOx removal efficiency can reach 88%. The NOx removal efficiency depended primarily on ozone / NOx ratio, and the temperature of flue gas also had influence on the NOx removal efficiency. The optimum pH range is 5.6-6.3. After inspection by authoritative institutions, the quality of fertilizers is superior to national standards.

1 Introduction

Development of simple, low cost, high efficiency of desulfurization and denitrification technology is the trend of ultra-low emissions in the power plant flue gas. It is at full power plant denitrification alternative solutions. At the same time, many kinds of pollutant removal is power plant technology. Compared with the traditional removal techniques, and at the same time removing aims to reduce the SOx at a lower cost, NOx and other heavy metals. The technology test and mature in business step by step, at a power plant in the further market shows great potential in [1].

In the 1970s, simultaneous removal of sulfur dioxide and nitrogen oxides was the first proposed and developed technology based on desulfurization. The technology is designed to eliminate two or more contaminants in a reactor vessel. Simplification of the program reduces area and investment. But until now, most of the simultaneous removal systems are still being developed in the lab, and there are no demonstrations in the industry [2]. The following shows the different synchronization removal routes:

1) Electron beam irradiation [3-5]

Principles: The flue gas is irradiated by electron beam with 800keV-1MeV. O2, N2, H2O decompose to radicals. The radicals oxidize SO2 and NOx into sulfuric acid and nitric acid; the resulting acids react with NH3 into chemical salts.

Advantages: The removal efficiency of SO2 and NOx is high. The scheme is simple. The side product NH4NO3 and (NH4)2SO4 can be further utilized as fertilizer.

Disadvantages: High voltage DC power supply and electron accelerator are high-cost devices. It also demands severe safety protection measures for X ray radiation and liquid ammonia leakage. Side products are areoles and difficult to be removed.

2) Pulse corona[6,7]

Principles: Pulsed high voltage power discharge to generate high-energy electrons

Advantages: No electron accelerator and radiation protection is needed, which reduces the costs.

Disadvantages: Not mature.

3) Metal oxide catalysis [8]

Principles: Metal oxides are used as catalysts to remove the SO2 and NOx. The most used metal oxides are CuO/Al2O3.

Advantages: At 300-500°C, CuO impregnated on Al2O3 could achieve 95% remove efficiency of SO2 and 90% remove efficiency of NOx.

Disadvantages: Sulfates are common side product. It is difficult to regenerate the catalysts.

4) Carbon-based catalytic method

Principles: SO2 is absorbed and oxidized to sulfuric acid by carbon-based materials. The absorbents are then used as catalyst for selective catalytic reduction denitrification, which converts NH3 and NOx into N2.
Advantages: High efficiency, low investment, side products can be further utilized, catalysts are be recycled.

Disadvantages: Interaction between SO\(_2\) and NO\(_x\) are inevitable. NO\(_x\) could improve SO\(_2\) removal efficiency, but SO\(_2\) decreases the NO\(_x\) removal efficiency.

5) Complex absorption method\(^{(9,10)}\)

Principles: Fe\(_{2}\) + is added in alkaline or neutral solution to form complex, which can absorb NO\(_x\). The resulting nitrosyl iron complexes dissolve SO\(_2\), O\(_2\) to form other complexes.

Advantages: SO\(_2\) removal efficiency could be 99%, and NO\(_x\) removal efficiency could be above 60%.

Disadvantages: The regeneration of absorbent is difficult. The absorbent is disposable. The cost is high.

Recently, the oxidation/absorption simultaneous removal methods are promising because of high efficiency, high value of by-products\(^{(11,12)}\). The primary scheme of this technology is: NO is not soluble in water, and NO is firstly oxidized to NO\(_2\), N\(_2\)O\(_3\) by oxidation agents such as O\(_3\), H\(_2\)O\(_2\), Cl\(_2\), ClO\(_2\). NO\(_2\), N\(_2\)O\(_3\) could be solved in water and react with alkaline solution\(^{(13-15)}\).

The choice of oxidant is a hot issue. ClO\(_2\) is an alternative because of its low cost, it can also be used for mercury removal. However, some surveys have shown that O\(_3\) is the safest and simplest oxidant, but at a high cost. However, with the development of technology, the O\(_3\) generator is a proven and effective cost. O\(_3\) becomes a suitable candidate\(^{(16)}\).

Factors that affect O\(_3\) and removal of NO\(_x\) and SO\(_2\): include NO/O\(_3\) mole ratio, temperature, reaction time, absorbent.Jiang et al. Investigate the DeNO\(_x\) using ozone. The results showed that NO\(_3\) is the product of the low O\(_3\)/ NO mole ratio. NO\(_3\), N\(_2\)O\(_3\) is O\(_3\)/ NO mole ratio is greater than 1.0 when the product of NO\(_x\) removal efficiency is more than 90% of aerosols, NO\(_3\) concentration in the liquid also increases.

Wang et al\(^{(17)}\) analyzed the oxidation of NO\(_x\) by ozone, and indicated that there are 40 intermediates and 121 steps chemical reaction mechanism. The mechanism is testimony and proves experimental research. By comparing the O\(_3\) life cycle and reaction kinetics, the half-life of the O\(_3\) decomposition is 19.2 seconds for a typical flue gas temperature of 150°C, which is not achieved in the slowest O\(_3\) and 0.6 seconds. At typical flue gas temperatures, no oxidation efficiency is above 84%, and desulfurization efficiency may be 100% if wet scrubbing is achieved. The DeNO\(_x\) efficiency and the O\(_3\)/impossible 86.27% molar ratio are 0.9. The young sun Mok Heon-Ju Lee O\(_3\) is injected into the flue gas to oxidize, the sodium and sodium hydroxide solution is used to absorb nitrogen oxides, the nitrogen oxides are finally converted to nitrogen, and the nitrogen oxide removal efficiency can be as high as 95%. The desulfurization efficiency can reach 100%.

Although the oxidation/absorption method has many advantages, the oxidation/adsorption method still has some problems when simultaneously removing NO\(_x\) and SO\(_2\).\(^{(18)}\)

1) NO\(_x\) removal efficiency requires higher and more stringent environmental standards, which directly increases O\(_3\) consumption. The advantage is marginal compared to thyristor.

2) The by-products have low economic value. For example, the by-products are (NH\(_4\))\(_2\)SO\(_4\) and NH\(_4\)NO\(_3\) using ammonia as absorbents. The (NH\(_4\))\(_2\)SO\(_4\) and NH\(_4\)NO\(_3\) can be used as fertilizers. But these by-products are mixed with other ammonium salts, which impair the fertilizers’ quality. Some of these salts even release SO\(_2\) into the atmosphere directly, causing second pollution.

3) Zero-valent mercury is difficult to solve in water. But Hg\(_2\) + is easy to absorb water and other alkaline solutions. Currently, zero-valent mercury is usually oxidized to Hg\(_2\)\(^{+}\) and then removed. This process also requires O\(_3\), and the consumption of O\(_3\) increases. In addition, mercury is absorbed by alkaline solutions and is also a by-product of contamination.

All of these simultaneous removal techniques, oxidation/adsorption methods remove high efficiency and high value added by-products. Oxidation/absorption has also proven the industry. But there are also some issues that need to be solved before the application.

### 2 Mechanism

The organic catalytic desulfurization and denitrification technology is based on the oxidation/absorption method. It could remove NO\(_x\) and SO\(_2\) in a single tower. The mechanisms are as following:

#### 2.1 Absorption mechanism of SO\(_2\)

Wet desulfurization process are: the generality of the SO\(_2\) generation H\(_2\)SO\(_3\) after reaction with water. The traditional wet washing method using limestone, magnesium oxide and sulfuric acid reaction, the formation of sulfate. Organic catalytic oxidation using catalyst and sulfuric acid to form stable multivalent compounds. The inhibition of sulfuric acid decomposition of adverse reaction. Then the residual oxygen in flue gas to the compound oxide into sulfuric acid. Sulfuric acid and alkaline substances such as ammonia reaction, form (NH\(_4\))\(_2\)SO\(_4\). The process just like industrial fertilizer production. Involved in the reaction is as follows:

\[
\begin{align*}
\text{SO}_2 + \text{H}_2\text{O} \rightarrow \text{H}_2\text{SO}_3 \\
\text{H}_2\text{SO}_3 + \text{L} \rightarrow \text{L} \cdot \text{H}_2\text{SO}_3 \\
2\text{L} \cdot \text{H}_2\text{SO}_3 + \text{O}_2 \rightarrow 2\text{L} + 2\text{H}_2\text{SO}_4 \\
\text{H}_2\text{SO}_4 + 2\text{NH}_3 \rightarrow (\text{NH}_4)_2\text{SO}_4
\end{align*}
\]

L is the organic catalyst.

#### 2.2 NO\(_x\) absorption mechanism

The denitrification is alike to desulfurization. In the atmosphere of strong oxidation agents, NO is converted into HNO\(_2\). The catalysts integrate with HNO\(_2\) and form stable covalent compound, which inhibits the HNO\(_2\) decomposition reversely. The catalysts also contributes to
the oxidization of intermediates into HNO₃. The alkaline solution is added and reacts with HNO₃, forming NH₄NO₃. This process is the same as the production of NH₄NO₃ in fertilizer industrial. The reactions involved are as following:

\[
\begin{align*}
3\text{NO} + \text{O}_3 & \rightarrow 3\text{NO}_2 \quad (5) \\
\text{NO}_2 + \text{NO} & \rightarrow \text{N}_2\text{O}_3 \quad (6) \\
\text{N}_2\text{O}_3 + \text{H}_2\text{O} & \rightarrow 2\text{HNO}_2 \quad (7) \\
\text{HNO}_2 + \text{L} & \rightarrow \text{L-HNO}_2 \quad (8) \\
2\text{L-HNO}_2 + \text{O}_2 & \rightarrow 2\text{L} + 2\text{HNO}_3 \quad (9) \\
\text{HNO}_3 + \text{NH}_3 & \rightarrow \text{NH}_4\text{NO}_3 \quad (10)
\end{align*}
\]

3 Experimental

3.1 Experimental setup

The experimental device is a tower, which is a container for easy transportation. The flue gas is pumped directly from the gas pipeline of the power plant into the test unit. The tower’s maximum processing capacity is 100m³/h of flue gas. The tower is 3,400 mm high and has a diameter of Φ165.2×4. The packing is filled in the column. There is a dealer above the tower. There is a solution tank under the tower. The solution in the pond is recovered in the column by a pneumatic diaphragm pump. The PH circuit is deployed in the circuit and uses the PID to control the pH of the solution. The flue gas enters the column through the packed catalyst and is finally discharged into the atmosphere. The ozone generator is Qingdao Guoling CF-G-3-50. The ozone production rate was 50 g/hr. The ozone is mixed in the mixer before the column. The ozone generator uses oxygen as a raw material. The final ozone fraction is 10% and the other gas is oxygen. In the loop of the tower, ammonia is injected. The pH is controlled by injecting ammonia. The gas analyzer is ROSEMENT NGA2000. Filter the gas before the gas analyzer.

\[\eta_{SO_2} = \frac{\text{outlet}_{SO_2}}{\text{inlet}_{SO_2}} \times 100\% \quad (11)\]
\[\eta_{NO_x} = \frac{\text{outlet}_{NO_x}}{\text{inlet}_{NO_x}} \times 100\% \quad (12)\]

\[\eta_{SO_2}\] is the denitration efficiency; \[\text{outlet}_{SO_2}\] is the SO₂ concentration at the outlet of the tower; \[\text{inlet}_{SO_2}\] is the SO₂ concentration at the inlet of the tower.

\[\eta_{NO_x}\] is the denitration efficiency; \[\text{outlet}_{NO_x}\] is the NOx concentration at the outlet of the tower; \[\text{inlet}_{NO_x}\] is the NOx concentration at the inlet of the tower.

4 Results and discussion

4.1 Temperature

The removal of NOx in this test could be divided into two steps: the oxidization of NOx and the absorption of NOx. NOx is oxidized into NO and NO₂ by O₃ or hydrogen peroxide:

\[3\text{NO} + \text{O}_3 \rightarrow 3\text{NO}_2\]

The reactions of denitration in the tower can be divided in two series depending on catalysts:

The NO₂ reacts with water without catalysts:
$3 \text{NO}_2 + \text{H}_2\text{O} \rightarrow 2\text{HNO}_3 + \text{NO}$  \hspace{1cm} (14)

The NO2 reacts with water with catalysts:

$\text{NO} + \text{NO}_2 \rightarrow \text{N}_2\text{O}_3$  \hspace{1cm} (15)

$\text{N}_2\text{O}_3 + \text{H}_2\text{O} \rightarrow 2\text{HNO}_2$  \hspace{1cm} (16)

$\text{HNO}_2 + \text{L} \rightarrow \text{L} \cdot \text{HNO}_2$  \hspace{1cm} (17)

$2\text{L} \cdot \text{HNO}_2 + \text{O}_2 \rightarrow 2\text{L} + 2\text{HNO}_3$  \hspace{1cm} (18)

It can be seen that the oxidation and absorption of NOx is exothermic and the solubility increases with increasing temperature. Therefore a lower temperature favors the reaction. In order to verify the effect of temperature, electric heating is added around the flue gas duct and the tank. Fig 3 shows the influence of temperature on the removal efficiency.

Fig. 2 The scheme of the experimental setup (1. Condensation tank; 2. Suction fan; 3. Flue gas flow meter; 4. Mixer; 5. Ozone injection; 6. Flue gas inlet; 7. Circulation pump; 8. PH meter; 9. Liquid Flow meter; 10. Tank; 11. Tower.)

Fig 3. The influence of flue gas temperature on the denitration and desulfurization.
It can be seen from Fig. 3 and Fig. 4, lower temperatures favour denitrification. The tank temperature is more influential than the flue gas temperature. High flue gas temperatures reduce the production of NO₂. However, since the flue gas is well mixed with ozone and is far away from the water tank, there is enough time for the reaction. Therefore, there is almost no impact on the test results. When the tank temperature is higher than 60 ℃, the denitration efficiency is lowered.

4.2 Oxidizer/NOx ratio

By adjusting the flue gas flow rate, ozone flow rate and hydrogen peroxide flow rate, different ratios of NO and NOx in the flue gas can be achieved. Theoretically, the absorption mechanism is different at different NO and NO₂ molar ratios. So there are different deletion effects.

\[
\frac{O_3}{NO} = 0 - 0.5
\]

At a \( \frac{O_3}{NO} \) molar ratio of 0-0.5

NO₂ will increase with the increase of \( \frac{O_3}{NO} \) molar ratio. There is almost no NO₂ hydrolysis. The entire denitration efficiency will increase with the rise of NO₂ concentration.

\[
\frac{O_3}{NO} = 0.5 - 1
\]

At a \( \frac{O_3}{NO} \) molar ratio of 0.5-1.0

Almost all of the reactions in (1) - (4) occur in the column, and the reaction sequence depends on the chemical Gibbs energy. Reaction (II) is preferred and the remaining NO₂ will react with water. Some NO₂ cannot be absorbed into the water and there is some NO. When the NO₂/NO molar ratio is higher than 50%, the efficiency in the tank will increase and then decrease as the \( \frac{O_3}{NO} \) ratio increases. When NOx is completely converted to NO₂, NO₂ first reacts with water to form NO, and the remaining NO will react with the remaining NO₂, and then follow the reaction (II).

\[
\frac{O_3}{NO} \geq 1
\]

At a \( \frac{O_3}{NO} \) molar ratio ≥1

With the increase of oxidizer, the NOx in the flue gas is only NO₂, and there is also some unreacted ozone. NO₂ reacts with water, forming NO. NO is oxidized by ozone and generates NO₂. NO₂ then repeatedly react with water. In fact, the reactions of NOx and ozone are complicated. Wang et al indicated that at an ozone/NO ratio of 1.5 stoichiometrically, the NOx can be fully converted into nitrate, and completely removed.

The influence of \( \frac{O_3}{NO} \) ratio on the removal efficiency is shown in Fig. 5.

![Fig. 4 The influence of tank temperature on the denitrification and desulfurization](image)

![Fig. 5. The removal efficiency at varied O3/NO molar ratio](image)
is added to improve the absorption in the tower. The test is as following:

Table 2. Hydrogen peroxide and ozone two stage oxidization test

| Condition       | NO/ppm | NO2/ppm | NOx/ppm |
|-----------------|--------|---------|---------|
| Raw flue gas    | 143    | 0.4     | 143.4   |
| After hydrogen  | 16     | 75.2    | 101.2   |
| purity          |        |         |         |
| After ozone     | 0      | 51.3    | 51.3    |
| After purification | 0    | 23.2    | 23.2    |

The NOx in the raw flue gas is measured at the inlet of the tower without any addition of oxidizer. With the addition of hydrogen peroxide, the NOx concentration at the inlet of the tower reduces to 101.2 ppm due to some of the NO reacts with hydrogen peroxide directly into HNO3. With the addition of ozone, the NOx concentration at the inlet of the tower reduces to 51.3 ppm. The ozone is mixed lots of air due to the ozone generator. The NOx is diluted by air. In fact, the NOx is slightly lower than that of the addition of hydrogen peroxide.

This is because in the ozone, some NOx is oxidized into higher NOx.

It can be seen that the addition of hydrogen peroxide in the second stage could achieve the same effect as that of using the surplus ozone. It indicates that the surplus ozone in the tower improve the absorption process in the tower. The results of different oxidization methods are the same.

4.3 Viscosity of the catalysts

Four catalysts are selected for comparison. The four catalysts are entitled a, b, c, d. a is similar to b, and is prone to be mixed with water, and the liquid is milky. Catalyst c is difficult to be mixed with water, and there are obvious layers in the tower. So the water is pumped in the lower layer, the catalysts are not fully participated in the reactions, which negatively effects the denitration efficiency. Catalyst d shows a well mixing characteristic and a high denitration efficiency.

Catalyst a is well mixed in the operation. It could be pumped to the packing section in the tower. The pressure loss in the tower increases with the rise of the liquid circulation. The increase of the pressure even fluctuate the flue gas meter. By adding some surface active agents, the viscosity decreases and the system is at stable operation.

The actual amount of catalyst c in the operation is limited. The increase of pressure in the tower is not significant, the flow meter could be operated smoothly. But the primary circulation agent is water, lots of water is lost with the operation. So the water in the liquid decreases.

From the experiments, catalyst a shows the best physical characteristic, and achieves the highest denitration efficiency.

4.4 Catalyst ratio and L/G ratio on the denitration efficiency

L/G ratio greater impact on removal efficiency of pollutant. In the industry, minimize the liquid-gas ratio can reduce operating costs, if the pollutant removal efficiency can meet the requirement. Causing the absorption tower fulfill with saddle ring fillers, and the length of the tower reaches 3.4 m, the absorption liquid in the lower half of the tower may overflow to the internal face of the tower. Therefore, the value of the L/G in the test was not suitable for project applications directly. But the trends and comparison of different types of catalyst which still has referential.

With a NOx oxidization ratio of 44%-45% at the inlet of the tower, the influence of the catalyst ratio and L/G ratio in the solution are compared in Fig. 6-Fig. 8.
Catalyst a is used, the catalyst ratio is 20%, 25%, 30%, 40%, 50%. The results are listed in Fig. 6. As shown in Fig. 5, a lower or a higher catalyst ratio doesn’t benefit the denitration. At a low ratio, the catalyst could not stabilize HNO3; at a high ratio, less gaseous NOx is absorbed by the liquid. At a L/G ratio less than 40L/m³, the liquid with 40% catalyst has a better denitration. At a L/G ratio higher than 40L/m³, the solution with 25% catalyst shows the best denitration characteristic.

The effects of L/G ratio also can be seen from Figure 6. With L/G ratio increasing, liquid absorbing denitration efficiency gradually increasing, when the catalyst ratio was 25%, L/G ratio is 90L/m³, the denitration efficiency up to 77.7%, and concentration of NOx at the outlet of absorber was 50.8mg / Nm³. When the L/G ratio less than 50L/m³, as the L/G increase, the denitration efficiency increases; but continue to increase the L/G ratio, the denitration efficiency increases not obvious.

Fig. 7 shows the denitration efficiency versus to catalyst ratio and L/G ratio. Catalyst b is selected in the experiment. The catalyst b ratio is 33.3% and 40%. It can be seen that the denitration efficiency is higher at a lower catalyst ratio. With the increase of the L/G ratio, absorbing liquid denitration efficiency increases, when the catalyst ratio was 33.2%, liquid-gas ratio is 125L / m³, the highest 86.3% denitrification efficiency, this time absorber outlet NOx concentration was 49.2mg / Nm³.

Fig. 8 shows the denitration efficiency versus to liquid gas ratio. Catalyst c is selected in the experiment. The catalyst b ratio is 20% and 33.2%. At a liquid gas ratio less than 45L/m³, the catalyst with a ratio of 33.2 shows a higher efficiency than that of 20% catalyst ratio. At a liquid gas ratio higher than 50L/m³, the catalyst with a ratio of 20% shows a higher efficiency than that of 33.2% catalyst ratio.

It can be seen that the catalyst ratio is an important factor on the denitration efficiency. For catalyst a, at a liquid gas ratio lower than 40L/m³, the solution with 40% catalyst shows the highest denitration efficiency. At a liquid gas ratio higher than 40L/m³, the solution with a 25% catalyst ratio shows the best denitration ratio. For catalyst b, the solution with 33.3% catalyst ratio has a higher denitration efficiency than that of 40% catalyst ratio. For catalyst c, at a liquid gas ratio less than 45L/m³, the solution with 33.5 ratio shows a better denitration efficiency; at a liquid gas ratio higher than 50L/m³, the solution with 20% catalyst ratio shows the best denitration efficiency.

4.5 The pH value of absorbing liquid on the DeSNOX efficiency

The pH value of absorbing liquid directly affect the removal efficiency of pollutants and the ammonia escape. The lower pH value, the lower concentration of NH4+, which could reduce the removal efficiency of pollutants. On the contrary, high pH value may cause other side effects and could lead to ammonia escape.

In order to test the influence of PH value on the removal efficiency of pollutants, the PH value of absorption liquid was changed by control the injection amount of ammonia. 25 L absorbing liquid is injecting into the tower, including catalyst 5.25 L and 18.75 L water.

Fig. 9 the influence of PH value on the denitration efficiency

As shown in the figure 9 above, under the condition of difference L/G, high denitration efficiency are located at around 6.1, denitration efficiency increases with the increase of pH value at first, this may be due to higher concentration of NH4+ in the solution, which increases the chances contacted with HNO3. With the further increase of pH, denitration efficiency decreased gradually, this may because high concentration of NH4+; nitrous acid directly reacted with NH4+ to produce nitrite, which reduced the efficiency of denitration. Therefore, we suggest that the PH value of absorbing liquid should operate between 5.8 to 6.3.

5 Conclusions

The test using sulfanyl functional groups as catalyst was proposed to simultaneously remove NOx, SO2 from flue gas, flow rate of the absorption solution. In order to obtain the best DeSNOX efficiency, the flow rate of the flue gas, L/G, flue gas temperature, concentration of the catalyst, ozone/NOx ratio, solution PH value has been adjusted. Results show that the DeSO2 efficiency is very high, can reach to 99% stability. The lower temperature of the flue gas, the higher DeNOX efficiency, the best PH value operating range was 5.6-6.3. The best DeNOx efficiency can reach to 88%. In addition, in order to reduce the operating cost of oxidant, another oxidant, H2O2 was used to oxidize NO, and the DeNOx efficiency can reach to 68%. After inspected by authoritative organization, the quality of the fertilizer was better than the national standard in China.

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