

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

Zhipeng Xue¹, Hao Chen², Minmin Zhao²

¹PH.D ,Huadian Electric Power Research Institute, NO. 1 Xiyuan 10 Road, Xihu District, Hangzhou Zhejiang Province, 310030, China

²ENGINEER,Huadian Electric Power Research Institute, NO. 2 Xiyuan 9 Road, Xihu District, Hangzhou Zhejiang Province, 310030, China

Abstract. A method was proposed to remove NO_x 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 NO_x removal efficiency in this experiment, many parameters were changed. Such as flue gas flow, ozone / NO_x 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 NO_x removal efficiency can reach 88%. The NO_x removal efficiency depended primarily on ozone / NO_x ratio, and the temperature of flue gas also had influence on the NO_x 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 SO_x at a lower cost, NO_x 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 NO_x into sulfuric acid and nitric acid; the resulting acids react with NH₃ into chemical salts.

Advantages: The removal efficiency of SO₂ and NO_x 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 NO_x. 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 NO_x.

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 denitration, which converts NH₃ and NO_x into N₂.

Advantages: High efficiency, lost investment, side products can be further utilized, catalysts are be recycled.

Disadvantages: Interaction between SO₂ and NO_x are inevitable. NO_x could improve SO₂ removal efficiency, but SO₂ decreases the NO_x removal efficiency.

5) Complex absorption method^[9,10]

Principles: Fe²⁺ is added in alkaline or neutral solution to form complex, which can absorb NO_x. The resulting nitrosyl iron complexes dissolve SO₂, O₂ to form other complexes.

Advantages: SO₂ 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 NO₂, N₂O₃ by oxidation agents such as O₃, H₂O₂, Cl₂, ClO₂. NO₂, N₂O₃ could be solved in water and react with alkaline solution^[13-15].

The choice of oxidant is a hot issue. ClO₂ is an alternative because of its low cost, it can also be used for mercury removal. However, some surveys have shown that O₃ is the safest and simplest oxidant, but at a high cost. However, with the development of technology, the O₃ generator is a proven and effective cost. O₃ becomes a suitable candidate^[16].

Factors that affect O₃ and removal of NO_x and SO₂ include NO/O₃ mole ratio, temperature, reaction time, absorbent. Jiang et al. Investigate the DeNO_x using ozone. The results showed that NO₂ is the product of the low O₃ / NO mole ratio. NO₃, N₂O₅ is O₃ / NO mole ratio is greater than 1.0 when the product of NO_x removal efficiency is more than 90% of aerosols, NO₃ 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 O₃ life cycle and reaction kinetics, the half-life of the O₃ decomposition is 19.2 seconds for a typical boiler flue gas temperature of 150°C, which is not achieved in the slowest O₃ 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 O₃/impossible 86.27% molar ratio are 0.9. The young sun Mok Heon-Ju Lee O₃ 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 SO₂.^[18]

1) NO_x removal efficiency requires higher and more stringent environmental standards, which directly

increases O₃ consumption. The advantage is marginal compared to thyristor.

2) The by-products have low economic value. For example, the by-products are (NH₄)₂SO₄ and NH₄NO₃ using ammonia as absorbents. The (NH₄)₂SO₄ and NH₄NO₃ 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 SO₂ into the atmosphere directly, causing second pollution.

3) Zero-valent mercury is difficult to solve in water. But Hg²⁺ is easy to absorb water and other alkaline solutions. Currently, zero-valent mercury is usually oxidized to Hg²⁺ and then removed. This process also requires O₃, and the consumption of O₃ increases. In addition, mercury is absorbed by alkaline solutions and is also a by-product of contamination.

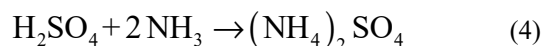
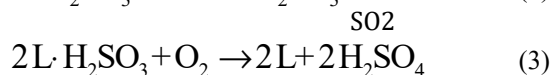
All of these simultaneous removal techniques, oxidation/adsorption methods remove high efficiency and high value added by-products. Oxidation/absorption has also proven the industry. But there are also some issues that need to be solved before the application.

2 Mechanism

The organic catalytic desulfurization and denitrification technology is based on the oxidation/absorption method. It could remove NO_x and SO₂ in a single tower. The mechanisms are as following:

2.1 Absorption mechanism of SO₂

Wet desulphurization process are: the generality of the SO₂ generation H₂SO₃ after reaction with water. The traditional wet washing method using limestone, magnesium oxide and sulfurous acid reaction, the formation of sulfate. Organic catalytic oxidation using catalyst and sulfurous acid to form stable multivalent compounds. The inhibition of sulfurous 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 (NH₄)₂SO₄. The process just like industrial fertilizer production. Involved in the reaction is as follows:

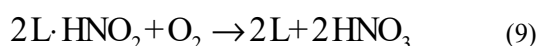
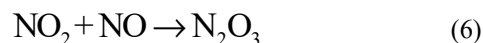
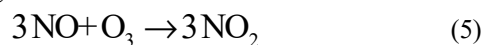


L is the organic catalyst.

2.2 NO_x absorption mechanism

The denitration is alike to desulfurization. In the atmosphere of strong oxidation agents, NO is converted into HNO₂. The catalysts integrate with HNO₂ and form stable covalent compound, which inhibits the HNO₂ decomposition reversely. The catalysts also contributes to

the oxidization of intermediates into HNO_3 . The alkaline solution is added and reacts with HNO_3 , forming NH_4NO_3 . This process is the same as the production of NH_4NO_3 in fertilizer industrial. The reactions involved are as following:



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 $\Phi 165.2 \times 4$. 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.



Fig. 1 The experimental setup

3.2 Materials and devices

The catalyst is oily. The viscosity is 14.5cP (20.8°C), 4.72 cP (60.1°C). The solubility in water is 8g/L, and the density is 0.942g/ml. The catalysts are added into the tower via the feeding pot before the experiment.

$$\eta_{\text{SO}_2} = \frac{\text{outlet}_{\text{SO}_2}}{\text{inlet}_{\text{SO}_2}} \times 100\% \quad (11)$$

$$\eta_{\text{NO}_x} = \frac{\text{outlet}_{\text{NO}_x}}{\text{inlet}_{\text{NO}_x}} \times 100\% \quad (12)$$

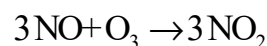
η_{SO_2} is the denitration efficiency; $\text{outlet}_{\text{SO}_2}$ is the SO_2 concentration at the outlet of the tower; $\text{inlet}_{\text{SO}_2}$ is the SO_2 concentration at the inlet of the tower.

η_{NO_x} is the denitration efficiency; $\text{outlet}_{\text{NO}_x}$ is the NO_x concentration at the outlet of the tower; $\text{inlet}_{\text{NO}_x}$ is the NO_x concentration at the inlet of the tower.

4 Results and discussion

4.1 Temperature

The removal of NO_x in this test could be divided into two steps: the oxidization of NO_x and the absorption of NO_x . NO_x is oxidized into NO and NO_2 by O_3 or hydrogen peroxide:

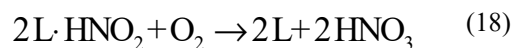
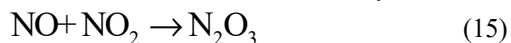


The reactions of denitration in the tower can be divided in two series depending on catalysts:

The NO_2 reacts with water without catalysts:



The NO₂ reacts with water with catalysts:



It can be seen that the oxidation and absorption of NO_x 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

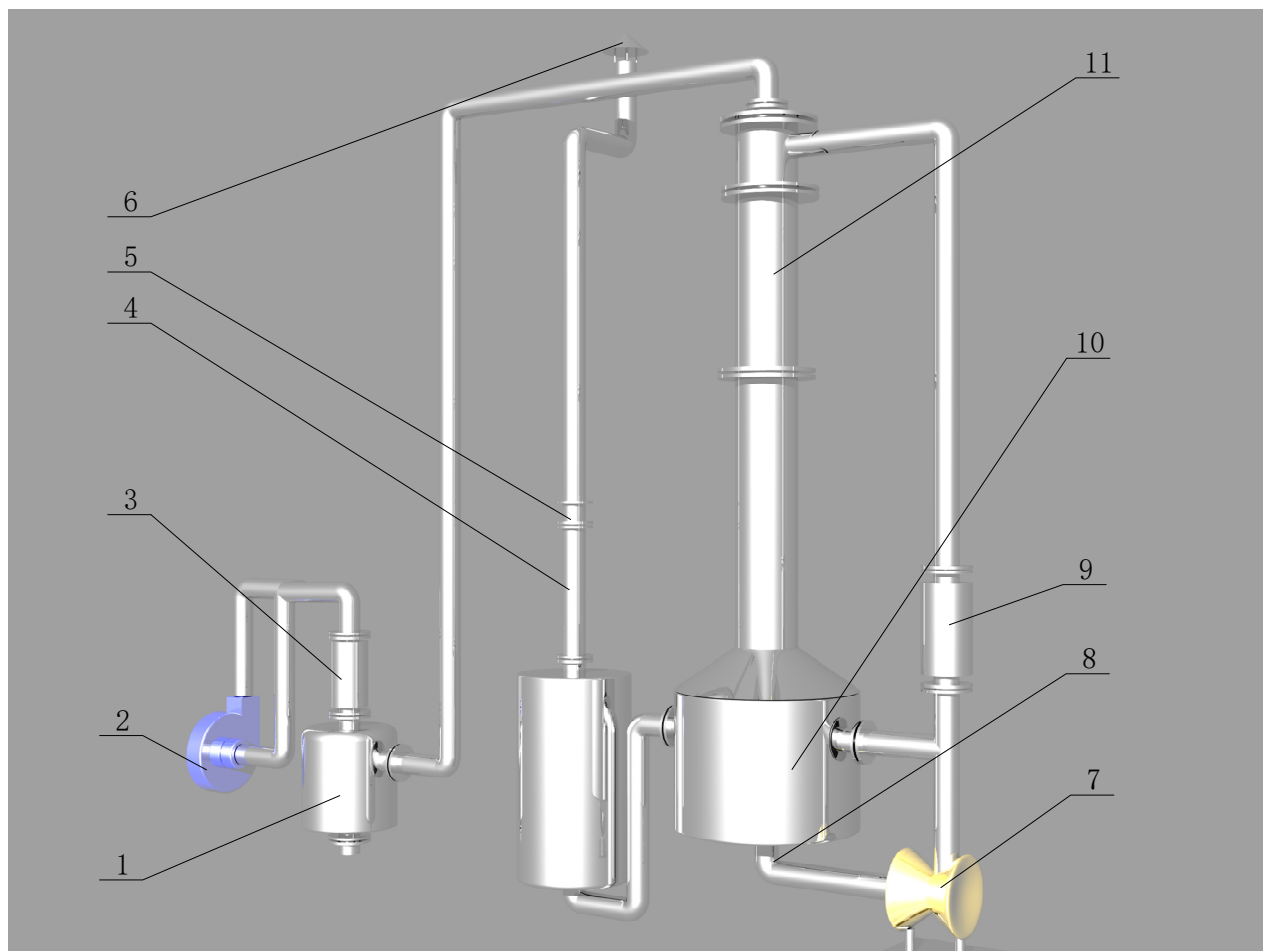


Fig. 2 The scheme of the experimental setup (1. Condensation tank; 2. Suction fan; 3. Flue gas flow meter; 4. Mixer; 5. Ozone injection; 6. Flue gas inlet; 7. Circulation pump; 8. PH meter; 9. Liquid Flow meter; 10. Tank; 11. Tower;)

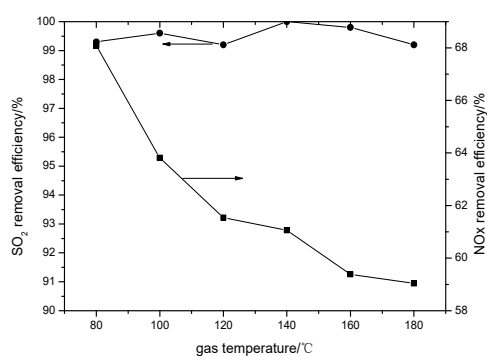


Fig 3. The influence of flue gas temperature on the denitration and desulfurization

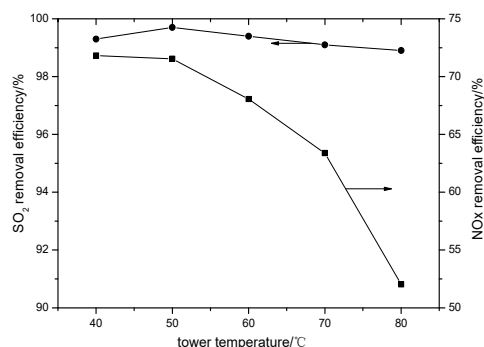


Fig. 4 The influence of tank temperature on the denitrification and desulfurization

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°C, the denitrification efficiency is lowered.

4.2 Oxidizer/NO_x ratio

By adjusting the flue gas flow rate, ozone flow rate and hydrogen peroxide flow rate, different ratios of NO and NO₂ 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.

$$O_3/NO = 0 \sim 0.5$$

At a O₃/NO molar ratio of 0-0.5

NO₂ will increase with the increase of O₃/NO molar ratio. There is almost no NO₂ hydrolysis. The entire denitrification efficiency will increase with the rise of NO₂ concentration.

$$O_3/NO = 0.5 \sim 1$$

At a O₃/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 O₃/NO ratio increases. When NO_x 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).

$$O_3/NO \geq 1$$

At a O₃/NO molar ratio ≥ 1

With the increase of oxidizer, the NO_x 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 NO_x and ozone are complicated. Wang et al indicated that at an ozone/NO ratio of 1.5 stoichiometrically, the NO_x can be fully converted into nitrate, and completely removed.

The influence of O₃/NO ratio on the removal efficiency is shown in Fig. 5.

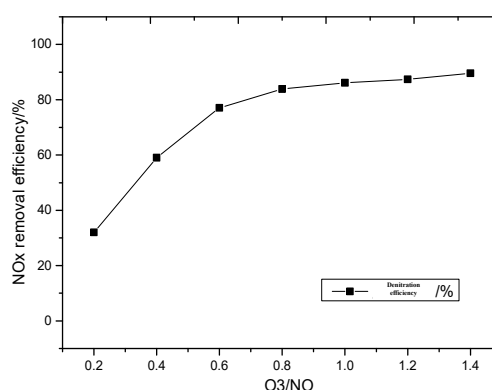


Fig. 5. The removal efficiency at varied O₃/NO molar ratio

The L / G ratio is maintained at 25 L / m³, the flue gas temperature is 135 °C, the catalyst concentration is 40%, the pH of the absorbent is maintained in the range of 6.1-6.2, and the total flow rate of the solution is 30 L. . As shown in Figure 5. When the O₃ / NO ratio is ≥ 1 , the denitrification efficiency is 90%, theoretically lower than 100%. This difference is due to ozone loss in other side reactions, which also reduces the impact of the catalyst.

At a O₃/NO ratio of 0-0.5, the NO is oxidized into NO₂ and then form into N₂O₃, but in the factual reaction, the NO/NO₂ is not in a ratio of 1, some ozone is lost in the side reactions, which impairs the oxidation.

At an O₃ / NO ratio of 0.5-1.0, NO is oxidized to NO₂ and then converted to N₂O₃ by catalysis. In the actual reaction, the NO / NO₂ ratio is equal to 1 or even higher than 1.0, and the denitrification efficiency is close to 80%. The result is also different from the theory, which is caused by the low NO_x concentration at the inlet.

At a O₃/NO ratio higher than 1.0, the experimental result nearly equals to the predication of theory. The ozone has a strong oxidizer. At a low NO_x concentration, the NO_x could be also oxidized to a higher state. The resulting HNO₃ reacts with the ammonia and form the stable nitrate, which benefits the oxidation reaction. However, there will be a large consumption of ozone, which increases the operation cost.

To further verify the absorption efficiency using ozone (O₃/NO ≥ 1) and reduce the ozone loss and cost. NO is oxidized by hydrogen peroxide at 350°C, and then ozone

is added to improve the absorption in the tower. The test is as following:

Table 2. Hydrogen peroxide and ozone two stage oxidization test

Flue gas condition	NO/ppm	NO ₂ /ppm	NO _x /ppm
Raw flue gas	143	0.4	143.4
After hydrogen peroxide	16	75.2	101.2
After ozone	0	51.3	51.3
After purification	0	23.2	23.2

The NO_x 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 NO_x 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 NO_x 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 NO_x is diluted by air. In fact, the NO_x is slightly lower than that of the addition of hydrogen peroxide.

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

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 NO_x 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.

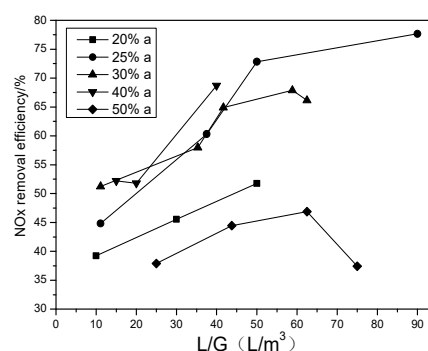


Fig. 6 Using catalyst a, the influence of liquid gas ratio on the denitration

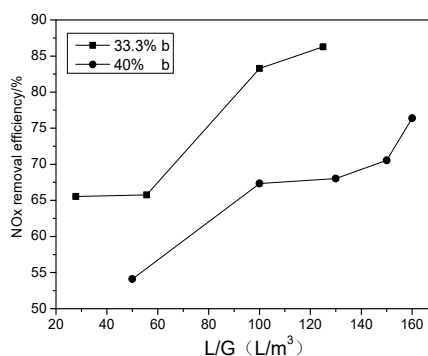


Fig. 7. Using catalyst b, the influence of liquid gas ratio on the denitration efficiency

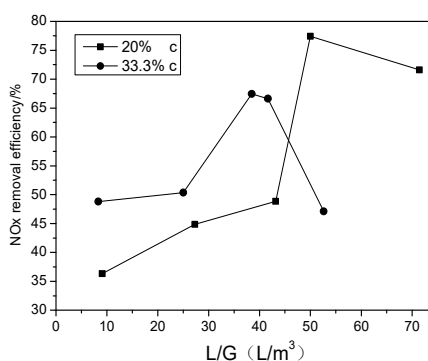


Fig. 8 Using catalyst c, the influence of liquid gas ratio on the denitration efficiency

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 HNO_2 ; at a high ratio, less gaseous NO_x is absorbed by the liquid. At a L/G ratio less than $40\text{L}/\text{m}^3$, the liquid with 40% catalyst has a better denetration. At a L/G ratio higher than $40\text{L}/\text{m}^3$, 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 $90\text{L}/\text{m}^3$, the denitration efficiency up to 77.7%, and concentration of NO_x at the outlet of absorber was $50.8\text{mg}/\text{Nm}^3$. When the L/G ratio less than $50\text{L}/\text{m}^3$, 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 $125\text{L}/\text{m}^3$, the highest 86.3% denitrification efficiency, this time absorber outlet NO_x concentration was $49.2\text{mg}/\text{Nm}^3$.

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 $45\text{L}/\text{m}^3$, 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 $50\text{L}/\text{m}^3$, 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 $40\text{L}/\text{m}^3$, the solution with 40% catalyst shows the highest denitration efficiency. At a liquid gas ratio higher than $40\text{L}/\text{m}^3$, 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 $45\text{L}/\text{m}^3$, the solution with 33.5 ratio shows a better denitration efficiency; at a liquid gas ratio higher than $50\text{L}/\text{m}^3$, the solution with 20% catalyst ratio shows the best denitration efficiency.

4.5 The pH value of absorbing liquid on the DeSNO_x 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 NH_4^+ , 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.

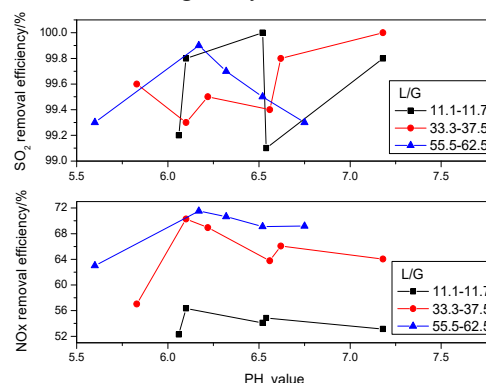


Fig. 9 the influence of PH value on the denitration efficiency

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 NH_4^+ in the solution, which increases the chances contacted with HNO_3 . With the further increase of pH, denitration efficiency decreased gradually, this may because high concentration of NH_4^+ , nitrous acid directly reacted with NH_4^+ 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 sulfinyl functional groups as catalyst was proposed to simultaneously remove NO_x , SO_2 from flue gas. flow rate of the absorption solution. In order to obtain the best DeSNO_x efficiency, the flow rate of the flue gas, L/G, flue gas temperature, concentration of the catalyst, ozone/ NO_x 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 DeNO_x efficiency, the best PH value operating range was 5.6-6.3. The best DeNO_x efficiency can reach to 88%. In addition, in order to reduce the operating cost of oxidant, another oxidant, H_2O_2 was used to oxidize NO, and the DeNO_x 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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