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# Simultaneous removal of SO<sub>2</sub> and NO<sub>x</sub> from biomass flue gas by ozone oxidation + wet scrubbing method

Zhenghui Xu<sup>1,2,3</sup>, Xiang Xiao<sup>1,3</sup>, Ping Fang<sup>1,3,\*</sup>, Jianhang Huang<sup>1,3</sup>, Haiwen Wu<sup>1,3</sup> and Dongyao Chen<sup>1,3</sup>

1. South China Institute of Environmental Sciences, the Ministry of Ecology and Environment, Guangzhou 510655, Guangdong China;

2. Key Laboratory of Poyang Lake Environment and Resource Utilization, Ministry of Education, School of Resources Environmental and Chemical Engineering, Nanchang University, Nanchang 330031, China;

3. The Key Laboratory of Water and Air Pollution Control of Guangdong Province, Guangzhou 510655, China;

corresponding author's e-mail address: fangping@scies.org

**Abstract:** In this work, factors affecting NO and SO<sub>2</sub> removal were systematically investigated, including O<sub>3</sub>/NO ratio (V/V), different oxidant types, NaOH and KMnO<sub>4</sub> concentrations. Results indicate that the degree of oxidation cannot be pursued too much and the best O<sub>3</sub>/NO ratio (V/V) is 0.5. The increased amount of KMnO<sub>4</sub> and NaOH promoted the removal efficiency of NO<sub>x</sub>. The key to the removal of NO<sub>x</sub> by KMnO<sub>4</sub> was mainly that when the KMnO<sub>4</sub> was 0.013 mol / L, the removal efficiency of SO<sub>2</sub> and NO<sub>x</sub> reached 100% and 93.75%, respectively. This indicates that the technology can not only efficiently remove NO and SO<sub>2</sub>, but also has a great application prospect.

## 1. Introduction

The use of biomass instead of fossil fuels to provide energy can alleviate the global warming problem that the international community faces, because biomass combustion does not cause a net increase in CO<sub>2</sub> emissions. <sup>[1-3]</sup>. At the same time, reducing the reliance on fossil fuels can also reset the energy used in biomass to produce renewable electricity, transportation and heat. <sup>[4,5]</sup>. In recent years, the general increase in the probability of using biomass for industrial production can also be attributed to the above reasons. Boiler is an important energy user in the industrial production process. At present, the substantial increase in biomass boilers has resulted in increased emissions of pollutants (such as NO<sub>x</sub>, SO<sub>2</sub>). For the development of control technologies for SO<sub>2</sub> and NO<sub>x</sub>, great progress has been made globally. Wet absorption is widely used in desulfurization because of its simple structure, high desulfurization efficiency and reliability. However, the low solubility of NO limits the appearance of high NO<sub>x</sub> removal efficiency. <sup>[6-8]</sup>. O<sub>3</sub> oxidation has the advantages of high oxidation efficiency without secondary pollution..

This experiment will study the optimal oxidation degree of O<sub>3</sub> oxidation combined with wet absorption. It is necessary to quantitatively oxidize a part of the NO in the flue gas to NO<sub>2</sub> in order to find a ratio of O<sub>3</sub> / NO (V / V) that is favorable for absorption, so as to reduce the oxidation cost and achieve a higher absorption effect. By determining the optimal reaction conditions, it provides



constructive guidance for the practical application of O<sub>3</sub> oxidation combined with wet absorption technology to remove SO<sub>2</sub> and NO simultaneously.

## 2. Experimental setup

### 2.1. Materials

NO (10.02%), N<sub>2</sub> (99.999%) and SO<sub>2</sub> (4.01%) purchased from Guangzhou Zhuozheng Gas Co., Ltd. were the three kinds of gases that need to be used. Sodium chlorite (> 78.00%, AR), sodium persulfate (≥98.00%, AR), potassium ferrate (≥ 98.58%, AR), potassium permanganate (≥ 99.50%, AR), all of them were got from Guangzhou chemical reagents.

### 2.2 Data analysis

$$\eta(\%) = \frac{NO_{int} - NO_{out}}{NO_{int}} \times 100\% \quad (1)$$

$$\eta(\%) = \frac{NO_{2int} - NO_{2out}}{NO_{2int}} \times 100\% \quad (2)$$

$$\eta(\%) = \frac{NOx_{int} - NOx_{out}}{NOx_{int}} \times 100\% \quad (3)$$

## 3 Results and discussion

### 3.1 Influence of O<sub>3</sub> / NO ratio (V/V)

Ideally, NO is converted into NO<sub>2</sub> as much as possible or dissolved in water with high-valent nitrogen oxides. However, the excessive amount of NO<sub>2</sub> will release NO after dissolving in water. Therefore, this study explored the effect of different O<sub>3</sub> / NO ratio (V/V) on NO<sub>x</sub> average removal efficiency, and ozone concentration changed from 40 ppm to 200 ppm. Figure 1 showed that when O<sub>3</sub> / NO ratio was changed from 0.2 to 0.5, the average removal efficiency of NO<sub>x</sub> gradually increased. The average NO<sub>x</sub> removal efficiency as high as 81.42% occurred when the ratio was 0.5. The continued increase in the ratio had a negative effect on the NO<sub>x</sub> removal efficiency. This may be because the increase in O<sub>3</sub> concentration caused more conversion of NO into NO<sub>2</sub>, which promoted the reaction (4), but the reaction (5) occurred when NO and NO<sub>2</sub> were present in the solution at the same time. This showed that higher ratio did not mean better outcome, there was an optimal range for the ratio. Based on comprehensive economic factors, it was known through experiments that the optimal O<sub>3</sub> / NO was about 0.5.

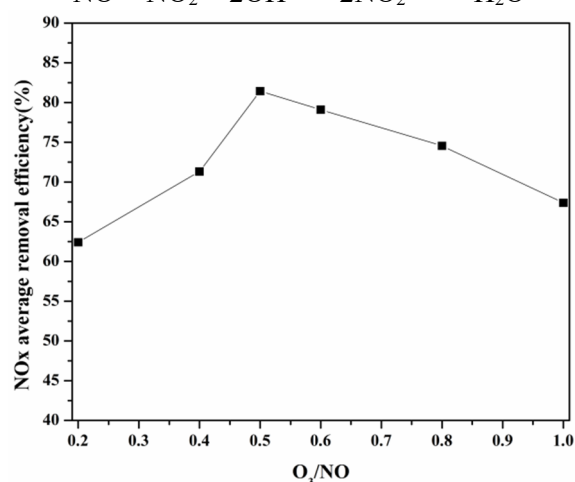
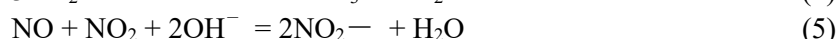
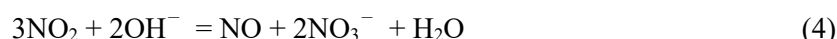


Figure 1. Effects of different O<sub>3</sub> / NO ratios (V/V) on NO<sub>x</sub> average removal efficiency. [KMnO<sub>4</sub>]= 0.006 mol / L, [NO]=200 ppm, [SO<sub>2</sub>]=150 ppm, [NaOH]= 0.025 mol / L.

### 3.2 Influence of the oxidant type

Adding different oxidants is an effective way to eliminate NO. In this study, four oxidants  $\text{Na}_2\text{S}_2\text{O}_8$ ,  $\text{NaClO}_2$ ,  $\text{K}_2\text{FeO}_4$  and  $\text{KMnO}_4$  were selected. Figure 2 was the effect of different oxidants on NOx removal efficiency. It can be seen that different oxidant solution has different removal effect on NOx due to the different oxidizing capacity. The capacity on the NOx removal of four oxidant solutions was  $\text{KMnO}_4 > \text{NaClO}_2 > \text{K}_2\text{FeO}_4 > \text{Na}_2\text{S}_2\text{O}_8$ , with the average NOx removal efficiencies being at 49.61%, 44.18%, 32.10% and 19.02%, respectively. The removal efficiency of  $\text{NaClO}_2$  at the beginning of the reaction was close to  $\text{KMnO}_4$ , but the stability of the whole reaction process was lower than that of  $\text{KMnO}_4$ . Therefore, considering various factors such as the cost of oxidant,  $\text{KMnO}_4$  was selected as the best additive.

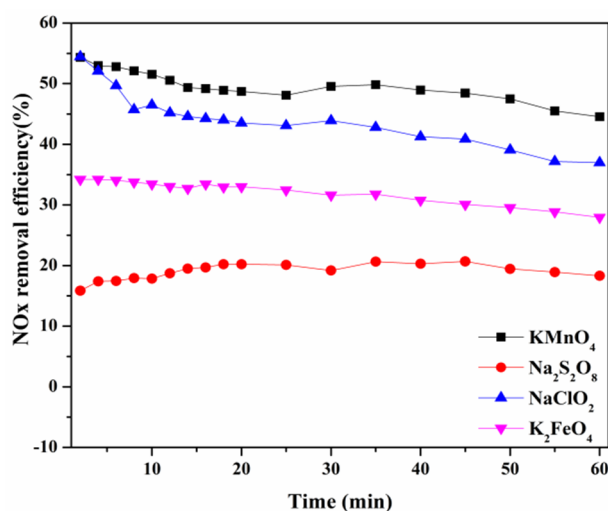


Figure 2. Effect of oxidant type.  $[\text{O}_3]=100$  ppm,  $[\text{SO}_2]=150$  ppm,  $[\text{NO}]=200$  ppm,  $[\text{KMnO}_4]=0.006$  mol / L,  $[\text{K}_2\text{FeO}_4]=0.005$  mol / L,  $[\text{NaClO}_2]=0.009$  mol / L,  $[\text{Na}_2\text{S}_2\text{O}_8]=0.004$  mol / L,  $[\text{NaOH}]=0$  mol / L.

### 3.3 Influence of NaOH

Figure 3 showed the effect of  $\text{KMnO}_4$  on NOx removal efficiency under the condition of changing NaOH concentration. It can be seen that the addition of NaOH had promoted the removal efficiency of NOx, and as the concentration of NaOH added increased, the removal efficiency of NOx increased, with the average NOx removal efficiencies being at 56.65%, 81.41%, 85.91% and 87.69%, respectively. When NaOH was 0.05 mol / L, the NOx removal efficiency can reach 88.89%, which can be attributed to the presence of  $\text{SO}_2$ . When increasing the concentration of NaOH, it can promote the conversion of  $\text{SO}_2$  to  $\text{Na}_2\text{SO}_3$  to a greater extent, thereby generating more sulfite which can react with  $\text{NO}_2$ . At the same time,  $\text{HNO}_2$  in solution can react with NaOH to generate  $\text{NaNO}_2$ , which can effectively prevent the decomposition of  $\text{HNO}_2$ . When NaOH concentration was more than 0.025 mol / L, the trend of increasing NOx removal efficiency gradually became slow. Therefore, the optimal NaOH concentration for subsequent experiments was chosen to be 0.025 mol / L. The efficiency of  $\text{SO}_2$  removal will not be discussed in subsequent experiments, because the removal efficiency was 100% throughout the experiment.

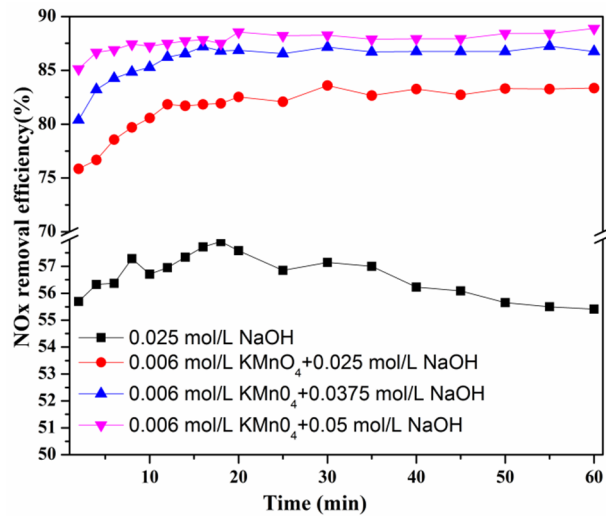


Figure 3. Effect of NaOH concentration. [NO]=200 ppm, [SO<sub>2</sub>]=150 ppm, [O<sub>3</sub>]=100 ppm.

3.4 Influence of KMnO<sub>4</sub> concentration

Figure 4 showed the results of different KMnO<sub>4</sub> concentrations on NO<sub>x</sub> removal efficiency. The increase of KMnO<sub>4</sub> concentration had a positive effect, with the average NO<sub>x</sub> removal efficiencies being at 79.44%, 84.44% and 92.86%, respectively. When KMnO<sub>4</sub> concentration was 0.013 mol / L, NO<sub>x</sub> removal efficiency obtained can reach 93.75% due to the increase of the collision probability. In conjunction with Figure 3, when KMnO<sub>4</sub> concentration exceeded 0.006 mol / L, the NO<sub>x</sub> increasing trend gradually slowed down. Therefore, the KMnO<sub>4</sub> concentration can be selected as 0.006 in practical applications.

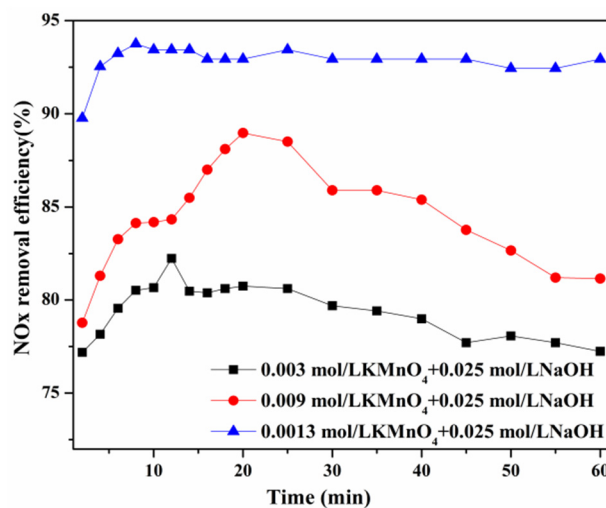


Figure 4. Effect of KMnO<sub>4</sub> concentration. [NO]=200 ppm, [O<sub>3</sub>]=100 ppm, [SO<sub>2</sub>]=150 ppm.

4. Conclusion

This study successfully proved a technology (ozone oxidation + wet scrubbing ) that can simultaneously remove SO<sub>2</sub> and NO<sub>x</sub>. After discovering that the SO<sub>2</sub> removal efficiency can reach 100%, the key factors affecting NO removal efficiency were explored. The conclusions were as following:

- (1) Ozone oxidation can significantly increase NO<sub>x</sub> removal. However, the degree of oxidation cannot be pursued too much and the best O<sub>3</sub> / NO ratio (V/V) is 0.5.

(2) The capacity on the NO<sub>x</sub> removal was KMnO<sub>4</sub> > NaClO<sub>2</sub> > K<sub>2</sub>FeO<sub>4</sub> > Na<sub>2</sub>S<sub>2</sub>O<sub>8</sub>.

(2) The increase of NaOH concentration will lead to an increasing trend of NO<sub>x</sub> removal efficiency. Under the condition of KMnO<sub>4</sub> concentration of 0.013 mol / L, the removal efficiency of NO<sub>x</sub> and SO<sub>2</sub> reached 93.75% and 100% respectively, which can prove that KMnO<sub>4</sub> played a key role.

### Acknowledgments

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