Simultaneous Removal of NO\textsubscript{x} and SO\textsubscript{2} by MgO Combined with O\textsubscript{3} Oxidation: The Influencing Factors and O\textsubscript{3} Consumption Distributions

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ABSTRACT: Simultaneous removal of NO\textsubscript{x} and SO\textsubscript{2} by MgO combined with O\textsubscript{3} oxidation was studied. The effects of the O\textsubscript{3}/NO molar ratio, oxidation temperature, and oxidation residence time on N\textsubscript{2}O\textsubscript{5} decomposition and O\textsubscript{3} consumption distributions were systematically illustrated, which is of great significance for improving NO\textsubscript{x} removal efficiency and reducing O\textsubscript{3} consumption in practical application. When the O\textsubscript{3}/NO molar ratio was greater than 1.0, the highest N\textsubscript{2}O\textsubscript{5} yield was achieved at 90 °C. The N\textsubscript{2}O\textsubscript{5} removal efficiency reached 96.5% at an O\textsubscript{3}/NO molar ratio of 1.8. The oxidation temperature increased from 90 to 130 °C, resulting in the decrease of N\textsubscript{2}O\textsubscript{5} yield, the improvement of O\textsubscript{3}-ICC (O\textsubscript{3} invalid cycle consumption) caused by N\textsubscript{2}O\textsubscript{5} decomposition, and the decrease of NO\textsubscript{x} removal efficiency from 96.5 to 76%.

1. INTRODUCTION

Sulfur dioxide (SO\textsubscript{2}) and nitrogen oxides (NO\textsubscript{x}) are recognized as the major pollutants causing environmental problems, such as acid rain and photochemical smog. With the NO\textsubscript{x} emission standards becoming more and more strict, numerous methods have been developed for NO\textsubscript{x} removal, such as selective catalytic reduction (SCR), activated carbon method, and ozone oxidation–absorption method. Among them, the ozone oxidation–absorption method has attracted great interest because of its advantages of simultaneous removal of SO\textsubscript{2} and NO\textsubscript{x} with high efficiency and low cost.

Huge emissions of iron ore sintering flue gas from steel industry with high gas flow amounts and low temperature (120–180 °C) are a great concern, and many efforts have been devoted to the development of denitrification technologies. In comparison to other denitrification strategies such as SCR and activated carbon method, ozone oxidation–absorption technology is more suitable because of its application temperature and operation convenience. In the ozone oxidation combined with wet flue gas desulfurization (WFGD), NO is oxidized into NO\textsubscript{2} or N\textsubscript{2}O\textsubscript{5} by injecting O\textsubscript{3} in front of the wet spray tower. Subsequently, NO\textsubscript{3} and SO\textsubscript{2} in the spray tower are simultaneously removed.

The WFGD include limestone–gypsum, ammonium, MgO, and double-alkali methods. The magnesium base WFGD has been reported to possess various advantages, such as low investment, high removal efficiency of SO\textsubscript{2}, and high comprehensive utilization value of byproducts. Additionally, it has been reported that MgO slurry reacts with SO\textsubscript{2} in the spray tower to produce MgSO\textsubscript{3} which promotes NO\textsubscript{x} removal. Accordingly, it would be feasible to use magnesium base WFGD combined with ozone oxidation to accomplish the simultaneous removal of NO\textsubscript{x} and SO\textsubscript{2} in flue gas.

In the ozone oxidation–absorption technology, the removal efficiency of NO\textsubscript{x} highly relies on the composition of NO\textsubscript{x}...
(solubility in water: 4 NO, 0.032 g/dm³; NO₂, 213.0 g/dm³; N₂O₅, 500.0 g/dm³).⁴³ Compared with NO₂, N₂O₅ is easier to be removed in the spraying system but needs more O₃ for its generation.⁴⁷,⁴⁸ Wang et al.⁴⁹ reported the reaction parameters including oxidation temperature, oxidation residence time, and adsorption and MgSO₃ addition on N₂O₅ yields and denitrification efficiency. Sun et al.⁵⁰ studied the effects of the pH value, initial SO₂ concentration, MgO concentration, and other operating parameters on the NO₂ removal efficiency. Shen et al.⁵¹ presented a novel magnesium-based WFGD process in which sodium thiosulfate was used to inhibit the oxidation of the desulfurization byproduct. However, these reports paid little attention to the decomposition of N₂O₅ at high temperatures and the consumption distribution of O₃ during the whole process. To study the decomposition of N₂O₅ and the consumption distribution of O₃ in the oxidation process, it is beneficial to improve NO₂ removal efficiency and reduce O₃ consumption.

In this paper, the effects of the molar ratio of O₃/NO, oxidation temperature, oxidation residence time, and adsorption operation parameters (pH value, initial concentration of SO₂ and MgSO₃ addition) on N₂O₅ yields and denitrification efficiency were studied. Additionally, the distribution of O₃ consumption during the whole process was particularly concerned.

2. RESULTS AND DISCUSSIONS

2.1. N₂O₅ Yield and O₃-ICC Ratio. 2.1.1. Factors Influencing the N₂O₅ Yield. The solubility of N₂O₅ is much higher than those of NO and NO₂, and the yield of N₂O₅ greatly influences the absorption efficiency of NO. The molar ratio of O₃/NO, the oxidation temperature, and the oxidation residence time are the key factors to determine the N₂O₅ yield. Considering the practical working conditions of the sintering process, the oxidation residence time was chosen to be 1.2 s. The influence of the O₃/NO molar ratio and oxidation temperature on N₂O₅ yield was studied with the oxidation residence time of 1.2 s, and the results were illustrated as Figure 1.

![Figure 1](https://example.com/figure1.png)

**Figure 1.** Yield of N₂O₅ under different O₃/NO molar ratios and temperatures (conditions: [NO], 200 ppm; [SO₂], 500 ppm; [O₂], 16%; gas flow rate Q, 6 L/min; and oxidation residence time = 1.2 s).

It can be seen that N₂O₅ was not formed when O₃/NO ≤ 1, and NO is gradually oxidized to NO₂, as shown by R1. The reaction of R1 is very fast, and it is the main reaction step when O₃/NO ≤ 1.⁴⁶,⁴⁷ Further increase of the O₃/NO molar ratio led to the generation N₂O₅ yield, which was mainly ascribed to reactions as R2 and R3.

\[
\text{NO + O₃ = NO₂ + O₂} \quad (\text{R1})
\]

\[
\text{NO₂ + O₃ = NO₃ + O₂} \quad (\text{R2})
\]

\[
\text{NO₂ + NO₃ = N₂O₅} \quad (\text{R3})
\]

For the oxidation temperature, a similar phenomenon was observed in our previous research.⁵⁸ The results showed that the N₂O₅ yield first increased and then decreased with the increase of oxidation temperature, reaching the highest value at 90 °C for all O₃/NO > 1. When the temperature of gas was higher than 90 °C, the N₂O₅ yield decreased very rapidly. Especially at 180 °C, N₂O₅ generation was not detected. The decrease of N₂O₅ at the high-temperature region (110–180 °C) was mainly ascribed to reactions of R4 and R5. Moreover, our previous research showed that O₃ had strong oxidation selectivity for NO₂, the addition of SO₂ had a little effect on N₂O₅ yield and O₃ consumption.

\[
\text{N₂O₅ = NO₂ + NO₃} \quad (\text{R4})
\]

\[
2\text{NO₃ = 2NO₂ + O₂} \quad (\text{R5})
\]

Oxidation residence time is also an important factor influencing the N₂O₅ yield in oxidation reaction. Herein, the oxidation residence time was tested between 0.5 and 6.7 s to investigate its correlation with N₂O₅ yield. Considering the yield of N₂O₅, the operating cost, the O₃ escape, and the O₃/NO molar ratio was suggested to be within the range of 1.5–2.0. Therefore, the O₃/NO molar ratio of 1.8 was chosen. In Wang et al.’s research,⁵⁹ the O₃/NO molar ratio also chose a similar value (1.75). As shown in Figure 2, for all five oxidation residence times, the N₂O₅ yield decreased with increasing oxidation temperature. In addition, the oxidation residence time had different effects on the N₂O₅ yield at different oxidation temperatures. When the oxidation temperature was 90 and 110 °C, the N₂O₅ yield increased first and then decreased with the extension of oxidation residence time, and the N₂O₅ yield reached the highest value at 2.5 s. When the oxidation temperature was 130 and 150 °C, the N₂O₅ yield decreased as the oxidation residence time was prolonged. These results indicated that choosing a reasonable oxidation residence time at different oxidation temperatures is beneficial to promote the formation of N₂O₅.

2.1.2. Factors Influencing O₃ Invalid Cyclical Consumption. During the oxidation process, extra O₃ consuming processes include N₂O₅ decomposition, O₃ decomposition, and reaction with SO₂. In our previous research,⁵⁶ it was found that O₃ hardly decomposed when the oxidation temperature ranged from 90 to
150 °C, and SO2 hardly consumed O3. Therefore, in this temperature range, the research was mainly focused on the O3 extra consumption caused by the decomposition of N2O5 regardless of the decomposition of O3 and the O3 consumed by the reaction with SO2.

N2O5 decomposition mainly generates NO2, especially at high temperatures. The part of NO2 reacts with O3 to reproduce N2O5 again, leading to invalid cyclical consumption of O3. In this process, the main reaction is as R2, R3, R4, and R5. Hence, it is meaningful to investigate the proportion of O3 extra wasted by invalid cyclical consumption (O3-ICC), which was caused by N2O5 decomposition. Figure 3 showed the ratios of O3-ICC at different temperatures, which were calculated based on the concentrations of NO2, N2O5, and unreacted O3.

As shown in Figure 3, it was found that when O3/NO ≤ 1, no O3-ICC appeared. The ratio of O3-ICC increased with improvement of temperature and O3/NO molar ratio when O3/NO > 1. For an O3/NO molar ratio of 1.8, the oxidation temperature changed from 60 to 180 °C, and the O3-ICC ratio was increased from 0 to 38%. The results indicate that O3 was all consumed to generate NO2, and no O3 was wasted when O3/NO ≤ 1; the decomposition rate of N2O5 increased and consumed more O3 with increasing temperature when O3/NO > 1. Moreover, when the oxidation temperature was 180 °C, the decomposition of O3 also consumed part of O3. These results indicated that a lower oxidation temperature was beneficial to decrease O3-ICC.

Subsequently, the effect of oxidation residence time on the consumption ratio of O3-ICC in the oxidation process was also studied with the O3/NO ratio of 1.8 at 90, 110, 130, and 150 °C, respectively. As summarized in Figure 4, the O3-ICC ratio increased with improvement of oxidation residence time and temperature. When the oxidation residence time was 0.5 or 1.2 s, O3-ICC was not observed at 90 °C. Further increase of the oxidation temperature led to the dramatic increase of the O3-ICC ratio. When the reaction temperature was 130 °C, the O3-ICC ratio increased from 10 to 40% with oxidation time extension from 0.5 to 6.7 s. The experimental results showed that longer oxidation residence time led to more N2O5 decomposition and higher O3-ICC ratio. This phenomenon is more pronounced at especially high temperatures.

2.2. Removal Efficiency of NOx and the O3 Consumption Distributions. 2.2.1. Removal Efficiency of NOx

After the oxidation process, the flue gas was introduced into the spraying tower for absorption. As shown in Figure 5, for O3/NO ≤ 1.0, the removal efficiency of NOx increased with the improvement of the molar ratio of O3/NO and different oxidation temperatures led to little change of the NOx removal efficiency. When O3/NO was >1.0, the NOx removal efficiency increased with the increase of the molar ratio of O3/NO and decreased with the improvement of the oxidation temperature. NOx removal efficiency increased from 43 to 97% with the improvement of the O3/NO molar ratio from 1.0 to 1.8 at 90 °C. The NOx removal efficiency increased from 43 to 76% when the molar ratio of O3/NO was changed from 1.0 to 1.8 at 130 °C.

Studies revealed that when O3/NO ≤ 1.0, the absorbed NOx were NO and NO2, and NO2 was more easily absorbed than NO. Therefore, the removal efficiency of NOx increased with the improvement of O3/NO molar ratio. The oxidation products were NO2 and N2O3 when O3/NO > 1.0. Considering N2O3 is more easily absorbed than NO2, removal efficiency of NOx increases with the increase of the concentration of N2O3. With the improvement of oxidation temperature, the decomposition of N2O3 was accelerated, and the N2O3 yield decreased, and NOx removal efficiency accordingly decreased.

Besides, O3 is also an atmospheric pollutant, and whether there is O3 escaping after absorption is a concern in the industrial applications. Hence, the amount of O3 escaping after absorption was analyzed at different temperatures and molar ratios. As shown in Figure 6, O3 escape gradually appeared in the absorbed gas with the increase of the molar ratio. On the contrary, O3 escape decreased with the improvement of the oxidation temperature. The oxidation temperature at 90 °C gave the
highest $O_3$ escape value. When the oxidation temperature was 180 °C, no $O_3$ escape was detected within the full range of the $O_3$/NO molar ratio tested.

The reason for the $O_3$ escape was that $O_3$ was not completely consumed in the oxidation reaction tube and the spray tower. As the oxidation temperature increases, $O_3$ is consumed more in the oxidation reactor. Therefore, the $O_3$ entering the spray tower was reduced and the $O_3$ escape was reduced.

The oxidation residence time has a significant effect on the $N_2O_5$ yield and thus on NOx removal efficiency. Hence, the corresponding research was conducted with an oxidation temperature at 130 °C. As shown in Figure 7, extension of the oxidation residence time led to the decrease of NOx removal efficiency when $O_3$/NO > 1.0. At $O_3$/NO = 1.8, the removal efficiencies of NOx were 96, 76, and 60% with the oxidation residence time being 0.5, 1.2, and 2.5 s, respectively.

Meanwhile, reducing the oxidation residence time will lead to increasing the amount of $O_3$ entering the spraying tower, thus affecting the amount of $O_3$ escape. Hence, $O_3$ escape after absorption was studied at different oxidation residence times and $O_3$/NO molar ratios. As shown in Figure 8, when the oxidation residence time was 0.5, 1.2, and 2.5 s, $O_3$ began to escape with the $O_3$/NO molar ratio increasing to 1.5, 2.0, and 2.5, respectively. The results revealed that when the oxidation temperature was 130 °C, shorter oxidation residence time led to less $O_3$-ICC, suggesting more $O_3$ entering the spraying tower. In the spraying tower, some $O_3$ would be consumed, and the remaining unreacted $O_3$ would escape from the spraying tower.

2.2.2. $O_3$ Consumption Distributions. In addition to the performance of desulfurization and denitrification, optimization of $O_3$ consumption is also of great importance in the ozone oxidation−absorption method. Great energy is consumed during the $O_3$ production process, and decreasing $O_3$ consumption is a main strategy to reduce the operation cost of the denitration device. In order to reduce $O_3$ consumption, it is necessary to establish an analytical method for $O_3$ consumption distributions under different conditions and to verify the influencing factors.

Based on the flue gas analysis after oxidation and absorption, the $O_3$ balance was calculated, and the $O_3$ distributions were classified into five parts (Figure 9): after oxidation, the product was generated as NO2 and $N_2O_5$, which could be analyzed at the outlet of the oxidation reactor. The oxidation product NO2 corresponds to equimolar $O_3$ consumption named as Part A.

The oxidation product $N_2O_5$ corresponds to the $O_3$ consumption classified as Part B (excluding $O_3$ consumption caused by decomposition of $N_2O_5$). As described above, some $O_3$ is extra wasted in the invalid cyclical consumption ($O_3$-ICC), which was caused by $N_2O_5$ decomposition. Herein, $O_3$-ICC was classified as Part C. The remaining $O_3$ enters into the spraying tower, and many reactions occurred in the tower through the reaction tube and spray tower, some $O_3$ would be consumed, and the remaining unreacted $O_3$ would escape from the spraying tower.

Figure 6. $O_3$ escape under different $O_3$/NO molar ratios and temperatures (conditions: [NO], 200 ppm; [SO2], 500 ppm; [O2], 16%; MgO slurry concentration C, 0.05 mol/L; initial pH of slurry, 9.25; gas flow rate $Q$, 6 L/min; and oxidation residence time = 1.2 s).

Figure 7. NOx removal efficiencies under different $O_3$/NO molar ratios and oxidation residence times (conditions: [NO], 200 ppm; [SO2], 500 ppm; [O2], 16%; MgO slurry concentration C, 0.05 mol/L; initial pH of slurry, 9.25; gas flow rate $Q$, 6 L/min; and gas temperature $T$, 130 °C).

Figure 8. $O_3$ escape under different $O_3$/NO molar ratios and oxidation residence times (conditions: [NO], 200 ppm; [SO2], 500 ppm; [O2], 16%; MgO slurry concentration C, 0.05 mol/L; initial pH of slurry, 9.25; gas flow rate $Q$, 6 L/min; and gas temperature $T$, 130 °C).

Figure 9. $O_3$ combined with the MgO oxidation−absorption process of the $O_3$ consumption distribution schematic diagram.
complicated steps, such as further oxidation of NO₂ to generate N₂O₅, the oxidation of SO₃²⁻ to produce SO₄²⁻, the oxidation NO₂⁻ to produce NO₃⁻, the oxidation of NO which was released during the absorption of NO₂, and so forth. It is very difficult to analyze every single small part of O₃ consumption for every reaction. Hence, the O₃ consumption in the spraying tower is defined as Part D. At last, the O₃ escaping from the spraying tower was classified as Part E.

After building the analytical method for O₃ consumption distributions, it was employed for the experiments conducted at 90, 110, 130, and 150 °C with the oxidation residence of 1.2 s and a O₃/NO molar ratio of 1.8. As shown in Figure 10, we found that the ratio of Part A and Part C increased with increasing oxidation temperature, while the ratios of Part B, Part D, and Part E decreased with increasing oxidation temperature. For the oxidation temperatures of 90 and 110 °C, the largest percentage was Part B. For the oxidation temperatures of 130 and 150 °C, the largest percentage was Part A. The ratio of Part E was the least in all four oxidation temperatures. This indicated that most O₃ was consumed, with very few O₃ escapes, and even no O₃ escape was detected at 130 and 150 °C.

With the increase of oxidation temperature, the decomposition rate of N₂O₅ was accelerated, the yield of SO₄²⁻ increased, and the concentration of NO₂ decreased. When the oxidation temperature changed from 90 to 150 °C, O₃-ICC increased, and O₃ entering the spray tower to participate in the reaction decreased. Therefore, the ratio of Part A and Part C increased and that of Part B, Part D, and Part E decreased. These results indicated that oxidation temperature is the key factor affecting O₃ consumption distribution.

2.3. Effects of Operation Parameters on Desulfurization and Denitrification. 2.3.1. Effect of the pH Value. Figure 11 showed the efficiencies in removing NOₓ and SO₂ at different pH values when the molar ratios of O₃/NO were 1.0 and 1.5. The slurry was continuously circulated in the spraying tower to absorb oxidized simulated flue gas.

It can be seen from Figure 11 that the pH value had little influence on MgO’s absorption of NOₓ and the removal efficiency fluctuation under different pH values was less than 5%. The trend of NOₓ removal efficiency at different pH values was similar when the molar ratio of O₃/NO was 1.0 and 1.5. As the pH changed from 9 to 8, the NOₓ removal efficiency was lowered. However, the pH further decreased from 8 to 4.5, the NOₓ removal efficiency did not change. In addition, as shown in Figure 12, the removal efficiency of SO₂ remains at 100% when the pH value was above 5.5. When the pH dropped to 4.5, the removal rate of SO₂ slightly decreased.

N₂O₅ is easily absorbed, so the decrease of NOₓ removal efficiency at different pH values is due to the decrease of NO₂ removal efficiency. It can be seen from following reaction R6 that the removal efficiency of NO₂ was related to the concentration of OH⁻. The concentration of OH⁻ decreased as the pH decreased and the NO₂ removal efficiency decreased. As the pH drops further, SO₃²⁻ converted to HSO₃⁻.
SO$_3^{2-}$ and HSO$_3^-$ would react with NO$_2$ in the slurry (R7 and R8). In this pH range (8–4.5), the positive effects of HSO$_3^-$ and the negative effects of OH$^-$ cancel each other out, and the NO$_2$ removal efficiency did not change with pH.

$$
2\text{NO}_2 + 2\text{OH}^- = \text{NO}_2^- + \text{NO}_3^- + \text{H}_2\text{O} \quad (\text{R6})
$$

$$
2\text{NO}_2 + \text{SO}_4^{2-} + \text{H}_2\text{O} = 2\text{NO}_2^- + \text{SO}_3^{2-} + 2\text{H}^+ \quad (\text{R7})
$$

$$
2\text{NO}_2 + \text{HSO}_3^- + \text{H}_2\text{O} = 2\text{NO}_2^- + \text{SO}_3^{2-} + 3\text{H}^+ \quad (\text{R8})
$$

2.3.2. Effect of Initial SO$_2$ Concentration. In the practical industrial applications, the concentration of SO$_2$ contained in flue gas fluctuates greatly. Therefore, it was necessary to study the initial concentration of SO$_2$. Figure 12 studied the effect of different initial SO$_2$ concentrations on the absorption of NO$_x$ by MgO.

As shown in Figure 12, when the molar ratio of O$_3$/NO was 0.5, 1.0, and 1.5, the removal efficiency of NO$_x$ increased with the increase of the initial concentration of SO$_2$, but the degree of increase was different. However, the NO$_2$ removal efficiency decreased when the molar ratio of O$_3$/NO is 1.8. It was found that SO$_3^{2-}$ could react with NO$_2$, thus promoting the absorption of NO$_x$ (R7). The concentration of SO$_2$ in the flue gas component is usually greater than that of NO. Therefore, the concentration of NO$_2$ largely determines the promoting effect of SO$_2$ on NO$_x$ absorption. NO$_2$ content in flue gas: O$_3$/NO molar ratio 1.0 > 0.5 > 1.5, SO$_2$ on the NO$_x$ absorption promotion effect decreased in turn. At the same time, SO$_3^{2-}$ generated by SO$_2$ will consume O$_3$ in the spraying tower and reduce the reaction between O$_3$ and NO$_2$, thus having a negative effect on NO$_x$ absorption (R9). When the molar ratio of O$_3$/NO was 1.8, the negative effect was more obvious.

$$
\text{SO}_3^{2-} + \text{O}_3 = \text{O}_2 + \text{SO}_4^{2-} \quad (\text{R9})
$$

2.3.3. Effect of Magnesium Sulfite. It has been reported that SO$_3^{2-}$ can react with NO$_2$ to promote the absorption of NO$_x$. Therefore, MgSO$_3$ with different concentrations was added to the MgO slurry to investigate its effect on the removal efficiency of NO$_x$. The results are shown in Figure 13. It is shown in this figure that the increase of the MgSO$_3$ concentration promoted the removal efficiency of NO$_2$. If there was no MgSO$_3$ in the slurry, the removal efficiency of NO$_2$ was 42.5%. While the MgSO$_3$ concentration increased to 100%, the removal efficiency increased to 79.5%. However, the promotion of MgSO$_3$ cannot be sustained. This is because the SO$_3^{2-}$ ions in the slurry are consumed by NO$_2$ and O$_3$ (R7 and R10). Therefore, as the concentration of MgSO$_3$ increased, the time for promoting NO$_2$ removal efficiency was prolonged.

$$
2\text{SO}_3^{2-} + \text{O}_3 = 2\text{SO}_4^{2-} \quad (\text{R10})
$$

3. CONCLUSIONS

The factors influencing the denitrification performance of MgO and the consumption distribution of O$_3$ were studied, including the O$_3$/NO molar ratio, oxidation temperature, oxidation residence time, initial concentration of SO$_2$, pH value, and addition of MgSO$_3$. Oxidation temperature and residence time affect N$_2$O$_5$ yield, O$_3$ consumption distribution, and NO$_x$ removal efficiency. Under the conditions of the O$_3$/NO molar ratio being 1.8 and oxidation residence time being 1.2 s, the highest N$_2$O$_5$ yield was achieved at 90 °C without O$_3$-ICC consumption and the NO$_x$ removal efficiency was 96.5%. When the oxidation temperature was improved to 130 °C, the O$_3$ consumption percentage corresponding to NO$_2$ was the largest, and the O$_3$-ICC ratio increased to 21.94%, and the NO$_x$ removal efficiency decreased to 76%. Nevertheless, when the oxidation residence time was shortened to 0.5 s, the O$_3$-ICC ratio decreased to 10%, and the NO$_x$ removal efficiency increased to 95.5%. When the O$_3$/NO molar ratio was 1.8, improvement of the SO$_2$ concentration led to the decrease of the NO$_x$ removal efficiency, revealing an inhibition effect. Besides, adding MgSO$_3$ to the MgO slurry can promote the absorption of NO$_2$ due to the reaction of NO$_2$ and SO$_3^{2-}$.

4. EXPERIMENTAL SECTION

4.1. Experimental System. A schematic diagram of the experimental setup for the simultaneous removal of NO$_x$ and SO$_2$ using MgO slurry-combined ozone oxidation is shown in Figure 14. The system included an ozone generator, gas supply system, oxidation reactor, spraying tower, and gas analysis system.

In this paper, cylinder gas (Beijing Hua Yuan Gas Chemical Co., Ltd.) was used as the gas source, and the mass flow meter (Beijing Sevenstar Electronics Co., Ltd.) is used to control the flow of simulated flue gas. The flow rates of NO and SO$_2$ were controlled at 120 and 150 ml/min (273.15 K and 1
NO2 is further oxidized to N2O5, and two basic reactions occur, than that of R2 (K2: 3.5×1017, K3: 2.2×1030, temperature: 25 °C). The NO3 content is rarely. Therefore, we calculate the concentration of N2O5 by the conservation of nitrogen oxide.

4.2. Removal Efficiency. The NOx yield is calculated by eq 1, the N2O5 concentration is calculated by eq 2, and the N2O3 yield is calculated by eq 3, and [NOx] is the reactor inlet NO concentrations; [NO2]a, and [N2O5]a are the reactor outlet NO2 and N2O5 concentrations, respectively.

\[
\text{NO2 yield} = \frac{[\text{NO}_2]_{b}}{[\text{NO}]_{a}} \times 100\% \\
\text{N2O5 concentration} = \frac{[\text{NO}_5]_{b} - [\text{NO}_2]_{b}}{2} \\
\text{N2O3 yield} = \frac{2[N\text{O}_2]_{b}/[\text{NO}]_{a}}{100\%}
\]

The NOx and SO2 removal efficiencies are calculated by eqs 4 and 5, respectively. \([\text{SO}_2]_{a}\) is the reactor inlet SO2 concentration, \([\text{NO}]_{a}\) and \([\text{SO}_2]_{a}\) are the spraying tower outlet NOx and SO2 concentrations, respectively. The NOx in this paper only includes NO, NO2, and N2O, excluding other NOx (such as NO3 and N2O).

\[
\text{NOx removal efficiency} = \frac{[\text{NO}]_{a} - [\text{NO}]_{b}}{[\text{NO}]_{a}} \times 100\% \\
\text{SO2 removal efficiency} = \frac{[\text{SO}_2]_{a} - [\text{SO}_2]_{b}}{[\text{SO}_2]_{a}} \times 100\%
\]

4.3. O3 Consumption Distributions. The destination of O3 can be divided into five parts. The ratio of O3 consumption distribution can be calculated by eqs 6–10. Because of the N2O5 decomposition, extra O3 was wasted by invalid cyclical consumption (O3-ICC). \([\text{O}_3]_{a}\) is the reactor inlet O3 concentration, \([\text{O}_3]_{b}\) is the reactor outlet O3 concentration, \([\text{O}_3]_{c}\), is the spraying tower outlet O3 concentration.

\[
\text{O3 consumption ratio corresponding to NO2} = \frac{[\text{NO}_2]_{b}/[\text{NO}]_{a}}{100}\% \\
\text{O3 consumption ratio corresponding to N2O3} = \frac{3[N\text{O}_2]_{b}/[\text{NO}]_{a}}{100}\% \\
\text{O3-ICC ratio} = \frac{([\text{O}_3]_{b} - [\text{NO}_2]_{b} - 3[N\text{O}_2]_{b} - [\text{O}_3]_{b})}{[\text{O}_3]_{a}} \times 100\% \\
\text{O3 consumption ratio in the tower} = \frac{([\text{O}_3]_{b} - [\text{O}_3]_{a})/[\text{O}_3]_{a}}{100}\% \\
\text{O3 escape ratio} = \frac{[\text{O}_3]_{a}/[\text{O}_3]_{c}}{100}\% \\
\]

**ASSOCIATED CONTENT**

3 Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.9b02502.

Yield of NOx at different O3/NO molar ratios and temperature; O3 concentration after oxidation at different O3/NO molar ratios and temperature; and effect of residence time in the spray tower on NOx removal efficiency (PDF)
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