

Feasibility Analysis of the Sintering Flue Gas Oxidation Method for Denitrification Technology Route

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Abstract

With the vigorous development of China's iron and steel industry and the introduction of ultra-low emission policies, the emission of pollutants such as SO2 and NO^x has received unprecedented attention. At present, the commonly used denitrification methods include selective catalytic reduction (SCR), active coke, etc. As a newly developed denitrification technology, oxidation denitrification is not widely used, and the technical level is mixed, and there might be problems such as yellow smoke, secondary pollution and ozone escape in the practical application. In this paper, problems existing in the denitrification process of sintering flue gas oxidation are analyzed, and a 320 m² sintering machine is taken as an example. Comparing the denitrification technology of sintering industry, it could be seen that the denitrification technology route of oxidation method has low pollution, low cost and high comprehensive environmental benefits, and has greatly potential development.

Keywords

Sintering, Air Pollution, Denitration by Oxidation

1. Introduction

The frequent occurrence of haze weather has become the focus of our people's attention, nitrogen oxides (NO_X) emission is one of the main factors causing haze, which is a serious hazard to the environment and human health, NO_X is emitted into the atmosphere in addition to the haze that may be caused by ozone hole, acid rain and photochemical pollution and other atmospheric pollution problems [\[1\]](#page-9-0) [\[2\].](#page-9-1) The sources of NO_X can be divided into industrial sources, living sources and mobile sources, in which NO_x emitted from living sources and mobile sources is more dispersed and not easy to deal with, whereas NO_x emitted from industrial sources is more centralized and easy to control and deal with [\[3\],](#page-9-2) and it is known from the statistical results of China's Environmental Statistics Yearbook that the total amount of NO_X emitted from industry is much higher than that emitted from life, and there exists a lot of room for reduction, so that the reduction of the main way to reduce the emission of air pollutants is to control the emission of NO_X from the industry [\[4\]](#page-9-3) [\[5\]](#page-9-4) [\[6\].](#page-9-5) The iron and steel industry is the largest pollutant emitter, but the iron and steel industry does not have any mature application of denitrification technology at present [\[7\].](#page-10-0)

The iron and steel industry is the pillar industry of China's national economy [\[8\].](#page-10-1) In 2023, China's crude steel output was 1019 million tons, ranking the first in the world, accounting for more than 50% of the global total output. At the same time, the iron and steel industry is the key emission sources of air pollution, and the problems such as long industrial chain, complex process and huge resource consumption are difficult to solve [\[9\]](#page-10-2) [\[10\].](#page-10-3) In addition, environmental pollutants such as SO_2 , NOx , particulate matter, heavy metals and dioxins are also produced during the operation of the steel-making system [\[11\]](#page-10-4) [\[12\]](#page-10-5) [\[13\].](#page-10-6) The main pollutant emission of iron and steel industry has exceeded that of electric power industry and become the largest pollutant emission source of industrial sector [\[14\].](#page-10-7) In consequence, the relevant policies and standards issued by the government for the iron and steel industry become increasingly strict, and the pressure faced by the iron and steel industry is greater than ever before.

Sintering flue gases are of great interest in the processes of the iron and steel industry due to their complex properties such as large gas fluctuations, its complex nature such as huge gas fluctuations and temperature fluctuations and temperature fluctuations, high water and oxygen content. In order to meet the ultra-low emission standards, sintering flue gas purification technologies are emerging.

The denitrification technologies for sintering flue gas can be mainly classified as SCR, activated carbon technology, and ozone oxidation-absorption. Compared with the other two desulfurization technologies, Ozone oxidation denitrification has been increasingly used due to its advantages of synergistic multi-pollutant control, water conservation, and low investment in equipment.

Ozone oxidation with liquid phase absorption desulfurization and denitrification is actually divided into two steps to achieve the purpose of denitrification [\[15\],](#page-10-8) firstly using oxidants to oxidize NO to soluble high-valent $NO₂$ or $N₂O₅$, and then using the existing desulfurization equipment to achieve the reduction and absorption of high-valent nitrogen oxides through certain optimization [\[16\].](#page-10-9)

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The oxidation of NO to $NO₂$ can be achieved by a variety of oxidizing agents, such as commonly used ozone, chlorine dioxide, sodium chlorite, etc [\[17\].](#page-10-10) The oxidizing agent has a high utilization rate and good selectivity for NO, and the oxidation rate of NO can be close to 100% [\[18\],](#page-10-11) and at the same time, intermediate products can be used to efficiently remove the dioxin-like substances in the flue gas [\[19\].](#page-10-12) Compared with the selective catalytic reduction method and the activated carbon method, the ozone oxidation adsorption method has the advantages of high oxidation selectivity, high oxidation efficiency and no secondary by-products. This technology will deeply oxidize NO to $NO₃$ and $N₂O₅$, thus significantly improving the denitrification efficiency. This technology can significantly improve the denitrification efficiency and successfully solve the problem that the traditional ozone oxidation and co-absorption technology cannot realize the ultra-low emission.

2. Ozone Oxidation Synergistic Liquid Phase Absorption Desulfurization and Denitrification Key Technology and Equipment

Gas-phase ozone (O_3) oxidation synergized with liquid-phase absorption for desulfurization and denitrification involves gas-phase Oxidation [\[20\],](#page-11-0) liquid-phase absorption and reduction of three processes: the first ozone is added to the flue mixer, so that the flue gas and ozone are fully mixed and oxidation reaction; flue gas into the absorption tower, in the liquid-phase absorption tower to achieve simultaneous absorption of SO_2 , NOx and other pollutants with high efficiency, O3/NO molar ratio of 1.5 - 2.0, denitrification products into the sintering process of reduction treatment.

Since the diffusion ability of N_2O_5 and SO_2 dissolved in the gas phase and water is comparable to the ability to dissolve in aqueous solution [\[21\],](#page-11-1) simultaneous high-efficiency desulfurization and denitrification can be achieved in the same absorption tower. SO_2 , heavy metal pollutants are also oxidized by O_3 , degraded to low toxicity or easy to absorb and remove the material.

The process flow is shown in **Figure 1**, O_3 is injected into the flue before the absorption tower through the injection mixer and mixed with the flue gas to oxidize NO_3 and N_2O_5 in depth. O_3 is added to the flue before the absorption tower through the mixer and mixed with the flue gas to oxidize NO to NO₃ and N_2O_5 , and then the flue gas enters into the absorption tower to realize simultaneous desulfurization and denitrification with the absorbing liquid. The purified flue gas is dusted and discharged, so that both sulfur dioxide and nitrogen oxides meet the control requirements of ultra-low emission. The wastewater is treated harmlessly and the nitrate component is reduced to N_2 in the blast furnace.

In comparison to SCR and activated carbon method, ozone oxidation-absorption shows the advantages of high oxidation selectivity, high oxidation efficiency, and no secondary byproducts [\[22\]](#page-11-2) [\[23\].](#page-11-3)

Figure 1. Ozone oxidation synergistic liquid phase absorption desulfurization and denitrification process flow.

3. Analysis of Denitration by Oxidation Method

3.1. Yellow Smoke Problem

The yellow smoke problem is the oxidation product $NO₂$ has not been effectively reduced or absorbed. Because $NO₂$ is a red-brown gas, obvious yellow color could appear when the emission concentration of $NO₂$ in the flue gas exceeds 30 mg/m³. This problem occurs in imperfect oxidation and denitrification engineering, which is for supervision and regulation [\[24\].](#page-11-4) Generally, the oxidation method of denitrification with yellow smoke phenomenon cannot achieve ultra-low emission levels, and it is also the "false denitrification" project that has been questioned the most, especially in the case of wet desulfurization, it is advisable to use peroxide method to oxidize NO to N_2O_5 , which could solve the problem of yellow smoke and actual emission exceeding the standard due to low absorption efficiency of $NO₂$ in conventional desulfurization system. It can also be solved by using the special denitrification agent absorption tower to greatly improve the absorption efficiency, and the oxidation denitrification method combined with semi-dry desulfurization has a low initial concentration of nitrogen oxides (<150 mg/m3), the route of initial oxidation to $NO₂$ could be adopted, under which conditions cheaper oxidants (such as chlorine dioxide [\[25\],](#page-11-5) sodium chlorite [\[26\]](#page-11-6) [\[27\],](#page-11-7) etc.) might be considered to reduce construction and operating costs.

3.2. Secondary Pollution of Absorbed Products

Nitrate has a positive effect on the application of wet desulfurization gypsum and semi-dry desulfurization ash in building materials' industry, improving the thermal insulation capacity of the material, and applied to other occasions to determine whether there is secondary pollution, nitric acid and nitrite in wet desulfurization wastewater if improper disposal will pollute the water body, the desulfurization wastewater of sintering machine using such denitrification technology should not be used for slag washing. It can be added to a mixture for consumption and utilization. From the perspective of laboratory irrigation mechanism analysis and practical engineering application, most of the decomposition products of nitric acid and nitrite carried in wastewater in the sintering machine production process are nitrogen, which does not cause the initial concentration of nitrogen oxides to be enriched and increased, and the secondary pollution problem can be completely avoided and solved.

3.3. Ozone Escape Pollution Problem

The greatest risk of ozone escape lies in the ozone generator workshop rather than the flue gas, and the ozone that does not participate in the reaction will quickly decompose into oxygen in the high humidity reducing atmosphere in the desulfurization tower, which theoretically does not produce secondary pollution, unless there is serious flue gas bias in the desulfurization tower design. Therefore, under the condition that excessive ozone injection does not cause secondary pollution, the problem of large fluctuations in flue gas parameters of the sintering machine can be effectively solved. After measuring several peroxide demonstration projects, ozone cannot be detected by ppm level instruments in the chimney position, while a very small amount of ozone can be detected by ppb level instruments, almost within the error range of 0 points. Ozone has been applied in China's water treatment industry for many years, the equipment is all domestic, and there are also perfect preventive measures for workshop escape.

3.4. Nitrogen Balance and Conversion Rate

In the process of denitrification by oxidation method, NO in the flue gas is first oxidized to $NO₂$ or $N₂O₅$, and all kinds of oxidants can achieve oxidation efficiency of more than 96%. There are two reactions of reduction and absorption of oxidation products in the desulfurization system, mainly reduction reaction. The difficulty of analysis of nitrogen element balance is that the part reduced to nitrogen cannot be measured. Therefore, in the actual nitrogen balance analysis process, the nitrogen element outlet is lower than the nitrogen element inlet, and this part of the difference is the part that is converted into nitrogen. The same problem exists in other denitrification technical routes. The proportion of nitrogen converted by different projects is also different, which is affected by the concentration of sulfur dioxide in the flue gas, the oxygen content, the temperature and the water content of the flue gas.

The process of ozone oxidation combined with semi-dry absorption reduction denitrification is to quickly and evenly mix ozone after the dust collector, oxidize the vast majority of NO in the flue gas to $NO₂$, and then reduce and absorb $NO₂$ in the semi-dry desulfurization tower, using sulfurized and absorption accelerator to produce N_2 and calcium nitrite, so as to achieve the purpose of denitrification. In the actual production process, the initial concentration of nitrogen oxides (calculated as $NO₂$) in a single sintering machine is 220 mg/m³ as an example. The smoke volume is 900,000 Nm³/h, and the desulfurization agent is about 1.8 t/h, and the desulfurization ash is about 2.5 t/h, and the denitrification agent is about 60 kg/h. The molar ratio of ozone to nitrogen oxides is 1.25, and under this condition, the main oxidation product is $NO₂$, which cannot form higher nitrogen oxides.

The whole process involves a reaction pathway as shown in [Figure 2](#page-5-0) below,

where oxygen generates ozone with oxidizing properties through an ionization reaction in the ozone generator, and ozone generates $NO₂$ with NO through a gas-phase reaction in the flue gas, the whole reaction being a stepwise reaction. The process was illustrated by the following Equations.

$$
O_3 + NO \rightarrow NO_2 + O_2 \tag{1}
$$

$$
2NO_2 + H_2O \to 2H^+ + NO_2^- + NO_3^- \tag{2}
$$

$$
H_2O + 2NO_3^- + 5SO_3^{2-} + acceleration \rightarrow 5SO_4^{2-} + N_2 + 2OH^-
$$
 (3)

The oxidized gas enters the semi-dry fluidizer bed desulfurization and denigration undergoes the fluidizer lime trapping and high pressure spray humidification and efficiency enhancement reaction. During this process, excess $O₃$ is reduced to O_2 on the surface of porous solid powder, NO_2 is finally formed into N₂ under the action of OH⁻, stabilizer and accelerator, and SO_3^{2-} in the liquid phase plays the main role of reaction. $NO₂$ in the fluidizer lime and gentrifying agent to participate in the subsequent chemical reaction, and ultimately most of the $NO₂$ is reduced to $N₂$, generating a very small amount of nitrate and nitrite. This mechanism approach has been verified by the mechanism test of Professor Tianle Zhu's [\[28\]](#page-11-8) research group (chromatographic verification as shown in [Figure 3](#page-6-0) below) and engineering practice (as shown in [Table 1,](#page-6-1) [Table 2,](#page-6-2) [Table](#page-6-3) [3\)](#page-6-3). As shown in [Figure 3\(a\),](#page-6-0) the peak time of N_2 under the corresponding column was determined by testing, and as shown in Figure $3(b)$, 20 - 40 min was the optimal time for denitrification, and with the increase of time, the denitrification capacity gradually decreased and less N_2 was produced. Comprehensive detection of high-temperature catalytic reduction NO^x was carried out on the flue gas at the tail of the sintering system, and the total amount of NOx was always maintained below 50 mg/nm³, and the desulfurization gypsum ion detection met the requirements of ultra-low emission NO^x emission.

Figure 3. NO₂ absorption process N migration.

Table 3. Remark information.

3.4.1. Nitrogen Entry

In the process of oxidation combined with semi-dry absorption and denitrifica-

tion, the input of nitrogen element comes from NO in the flue gas after the dust collector, and the total input nitrogen element (nitrogen is not calculated) is: (equations number)

$$
N = V * c * 14/46 = 48.7 kg/h
$$
 (4)

----V Smoke flow rate

----c Nitrogen oxide concentration

3.4.2. Nitrogen Outlet

The nitrogen outlet consists of three parts, which are the residual emission of nitrogen oxides in the flue gas, nitrogen generated by the reaction of $NO₂$ and denitrification agent, and calcium nitrite (calcium nitrate) in desulfurization ash. Due to the high concentration of nitrogen in the flue gas itself, the nitrogen part cannot be detected and can only be analyzed through balance.

3.4.3. Residual Nitrogen Oxides in Flue Gas

According to the field measurement, the average concentration of nitrogen oxides in the final emission of flue gas calculated as $NO₂$ is 15 mg/m³, so the nitrogen element of this part is calculated as:

$$
N = V * c * 14/46 = 3.65 kg/h
$$
 (5)

The initial total nitrogen ratio was a $1 = 3.65/48.7*100\% = 7.5\%$.

3.4.4. Calcium Nitrite in Desulfurization Ash

The calcium nitrate content of the desulfurization ash is less than 2%, and at 2%, an average of 2 tons of desulfurization ash is produced per hour, so this fraction of nitrogen is calculated as:

$$
N = G * c * 28/132 = 8.5 kg/h
$$
 (6)

The initial total nitrogen ratio is $a2 = 8.5/48.7*100\% = 17.5\%$.

3.4.5. Part of Reduction to Nitrogen

According to the decrement method, the nitrogen element of this part is:

$$
a3 = 1 - 7.5\% - 17.5\% = 75\%
$$

4. Comparison of Denitrification Technologies in the Sintering Industry (Taking 300 m2 Sintering Machine as an Example) (As Shown in [Table 4\)](#page-7-0)

Table 4. Comparison of denitration technology in sintering industry.

Continued

5. Conclusions

The above analysis shows that the perfect implementation of oxidative denitrification is a very suitable technical route for denitrification of sintering machine, compared with other technical routes, it has the great advantages of low cost, small secondary pollution, high comprehensive environmental benefits, and its problems have a reliable solution to the problems, the recent report of the National Air Pollution Causes Group has also clearly pointed out the shortcomings of the SCR technical route, and made it clear that ammonia escape has a great influence on the atmosphere. The recent report of the National Air Pollution Causes Group has also clearly pointed out the shortcomings of the SCR technology route, and made it clear that ammonia escape is a great danger to atmospheric particulate matter and ozone pollution.

With the requirement of ultra-low emission of sintering flue gas being put on the agenda, the traditional denitrification and desulfurization technologies have been difficult to meet the requirements. Through the summarization of this paper, it is found that O_3 oxidation combined with semi-dry method for denitrification of sintering flue gas is the main trend of industrial flue gas treatment application in the future. The process is in line with the sustainable development concept of energy saving and emission reduction. The technology has been successfully applied in the iron and steel sintering industry, and in the future, it can be further popularized and applied to the flue gas treatment of cement, glass, ceramics, non-electric coal-fired boilers and other non-electricity industries, which has a broad market application prospect.

Data Availability

The data used and/or analyzed during the current study are available from the corresponding author on reasonable request.

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Conflicts of Interest

The authors declare no conflicts of interest.

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