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Application of A High Efficiency Composite Denitration Agent in Ceramic Roller Kiln Flue Gas Treatment: A Case Study

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Abstract. With the increasing stringent environment pollution emission standards, the ceramic industry is facing a great challenge of NO_x control. The research and application of highly efficiency deNO_x technologies for ceramic roller kiln are benefit both to ceramic industry and environmental protection. Selective non-catalytic reduction (SNCR) is among the leading edge of the hotspot of NO_x control. However, most of the reported investigations were laboratory scale study, which were limited to representing the performance of SNCR denitration agent at real application. In this study, we compared the performance of a self-made denitration agent with traditional ones, and the effect of spraying rate and position on NO_x removal efficiency were investigated at a 20000 m²/h ceramic product line. Besides, a continuous experiment was carried out. By the work conducted in a real gas, a number of first-hand data focusing on SNCR technology for deNO_x of ceramic roller kiln and its possibility of coupling with existed wet absorption process were shared in this paper.

1. Introduction

With the increasing serious air pollution problem, China has proposed a 10% reduction of NO_x constraint index during the 12th Five Year Plan period. As the advancement of ultra-low pollutants emission of thermal power plants, the reduction of NO_x emission of ceramic industry is playing a more and more important role. In 2010, the Ministry of Environmental Protection promulgated and implemented the “Emission standard of pollutants for ceramics industry (GB25464-2010)” and revised it in 2014. In the amending list, the emission standards of particulate matter (PM), SO₂ and NO_x were revised to 30, 50 and 180 mg/m³ respectively. Meanwhile, with the continuous improvement of production technology and flue gas treatment technology in China’s ceramic industry, some provinces in China have issued stricter local emission standards for ceramic industry. Shangdong province has tightened the emission standards of PM, SO₂ and NO_x to 10, 35 and 80 mg/m³ for ceramic industry in key control areas. Other major ceramic producing areas are also tightening the emission standards for ceramic industry. Guangdong province now is revising its local standards. In the draft, the limits for emission concentration of PM, SO₂ and NO_x are 20, 30, 100 mg/m³ respectively. Therefore, it is very urgent and necessary to upgrade the NO_x control technology in ceramic industry.

The NO_x emission of ceramic industry mainly contains of fuel-NO_x and thermal-NO_x. Ceramic raw materials and fuels contain a certain amount of nitrogen components. Under high temperature



combustion conditions, these nitrogen components generate fuel-NO_x through thermal decomposition, oxidation and other ways. The production of fuel-NO_x is closely related to the content of nitrogen components. Previous studies show that when the content of nitrogen in ceramic raw materials exceeds 0.1%, the fuel NO_x in flue gas will dominate [1]. In the conventional temperature of ceramic roller kiln, fuel-NO_x is the main NO_x to control, which accounts for 90-98% [2].

At present, the flue gas deNO_x technology of ceramic kiln mainly includes SNCR and SCR technologies. Among them, SNCR technology has been widely used, especially in the flue gas terminal treatment. For example, a ceramic manufacturing enterprise in Guangdong province adopts SNCR technology in the spray drying tower and the final outlet NO_x emission concentration is controlled at 80 mg/Nm³ [3]. On the basis of urea wet denitrification technology, Fang et al [4,5] developed a wet multi-pollutant synergistic control technology by adding additives, which can achieve the synergistic removal of NO_x, SO₂ and heavy metals, and the removal rate of NO_x is more than 50%. However, at present, the SNCR technology used in the treatment of ceramic flue gas is mainly terminal treatment, but the research on firing process is relatively scarce. In this study, the experiment was carried out on a ceramic production line to investigate the application of SNCR technology on deNO_x of fire-belt process-control.

2. Materials and methods

2.1. DeNO_x agent and experimental site

In this study, the deNO_x agent was a self-made composite denitration agent, the main components were ammonia and H₂O₂, and small 50 ppm Na-containing additive was added to it.

The experimental site was located at a ceramic brick production line in a ceramic factory in Zhaoqing city, Guangdong Province. The production capacity of this line is 20000 m²/d, and the flow rate of flue gas was 20000 m³/h. The experiment was conducted at the roller hearth kiln fire-belt with the temperature of 850-1150 °C. The concentrations of flue gas pollutants were shown in Table 1. During the experiment, the initial concentration of NO_x was stabilized at 110-160 mg/m³.

Table 1. The concentrations of flue gas pollutant.

	O ₂	PM	SO ₂	NO _x	
Flue gas	14-16%	50-200	600-1000	80-200	
National standard *	18%	30	50	180	
	Pb	Cd	Ni	F	Cl
Flue gas	-0.20	-0.02	-0.75	0.5-5	5-30
National standard *	0.5	0.5	0.5	5.0	50

*Emission standard of pollutants for ceramic industry (GB 25464-2010, 2014 amending list)

2.2. Analytical methods

The concentration of NO, NO₂ and O₂ before and after denitrification were detected by Testo 350 flue gas analyzer (Germany). NO_x is the sum of NO and NO₂ and is calculated in terms of NO₂. To compared with relevant emission standards, the NO_x volume concentration (ppm) which directly measured by flue gas analyzer was converted to mass concentration at 18% standard oxygen content, the equation is as follows:

$$\text{NO}_x(\text{mg}/\text{m}^3) = \text{NO}_x(\text{ppm}) \times \frac{M_r}{22.4} \times \frac{21\% - \varphi_{st}}{21\% - \varphi_m} \quad (1)$$

Which, M_r - the relative molecule mass, 46 g/mol; φ_{st} - the standard volume content of oxygen, 18%; φ_m - the volume content of oxygen during the experiment, %.

NO_x removal efficiency is defined as follow:

$$\text{NO}_x \text{ Removal efficiency} = (\text{NO}_{x\text{in}} - \text{NO}_{x\text{out}}) / \text{NO}_{x\text{in}} \times 100\% \quad (2)$$

3. Results and discussion

A simple spraying gun was used in this study to investigate the effect of self-made composite denitration agent on NO_x removal comparing with common deNO_x agents such as urea and ammonia with the NSR of 1.2. The results are shown in figure 1.

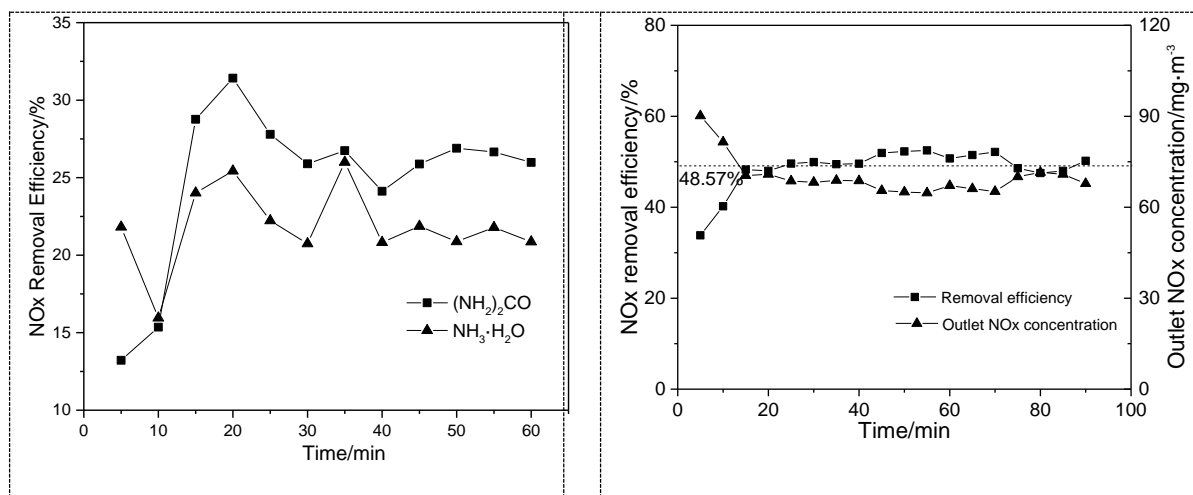


Figure 1. The NO_x removal efficiency of different denitrification agent.

When the injecting rate was 2.78 L/min and the injecting position was at A point, the outlet NO_x concentration remained below 90 mg/m³. After the beginning 20 min of reaction, the NO_x concentration basically stable at about 75 mg/m³. During 90 min consecutive continuous experiment, the average removal efficiency of NO_x was 48.57%, which was far more favorable than urea and ammonia, as shown in figure 1. The self-made composite denitration agent was composed of ammonia and Na-containing additive. The additive content of ammonia was same as above, and the concentration of Na-containing additive was 50 ppm. It can be seen in figure 1 the adding of Na-containing additive can get much higher NO_x reduction. The reason can be explained as that, when Na ions were added, a cyclic reaction of NaOH→Na₂O→NaO→NaOH would occur. During the reaction, a large number of OH groups would be produced, and NH₃ would be transformed into intermediate products NH₂ and NCO, thus promoting the removal of NO_x [6,7]. Thus, sodium metal salts promoted denitrification mainly by cationic denitrification.

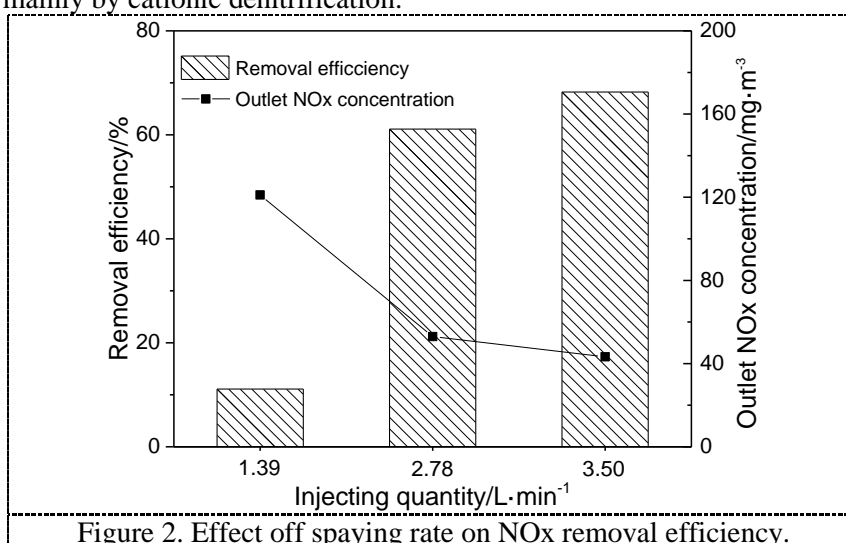


Figure 2. Effect off spaying rate on NO_x removal efficiency.

The effect of different spaying rate of the composite denitration agent on NO_x removal efficiency was studied. The results are shown in figure 2. As can be seen in figure 2, the removal efficiency of

NOx increased with the rising injecting rate of composite deNOx agent. As the injecting quantity was 3.50 L/min, the transient removal efficiency of NOx was close to 70%, the outlet concentration of NOx is about 40 mg/m³, which far below the relevant national and local NOx emission standard of ceramic industry. While considering the control of slip quantity of ammonia, the injecting quantity was set at 2.78 L/min.

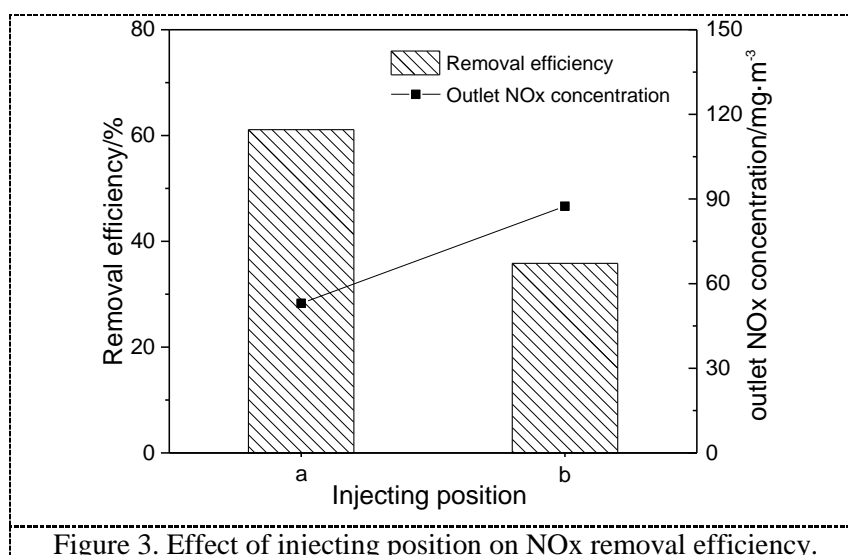


Figure 3. Effect of injecting position on NOx removal efficiency.

When the spraying rate was 2.78 L/min, the effect of changing the position of the composite deNOx agent injecting site was shown in figure 3. It can be seen in figure 3 that the injecting position of deNOx agent has a great influence on the removal efficiency of NOx. The NOx removal efficiency was much higher when spraying from position A than from position B. When composite denitration agent was sprayed at position A, the NOx removal efficiency was above 60%, and the outlet NOx concentration was 53 mg/m³. Because of the same type of spraying gun and same spraying quantity, it can be deduced that different injecting position mainly affected the reaction temperature of deNOx agent and NOx, which means the temperature is the main influencing factor of denitrification.

4. Conclusion

In this study, a high efficiency composite denitration agent was used in a ceramic production line for NOx control. The main conclusions are shown as follows:

- (1) During 90 min consecutive experiment, the average NOx removal efficiency was 48.57%, and the outlet NOx concentration was basically stable at about 75 mg/m³, which was far more favorable than urea and ammonia.
- (2) The spraying rate and position can both affect the deNOx efficiency. The removal efficiency of NOx increased with larger denitrification agent amount. Changing the position of composite deNOx agent affected the reaction temperature, which affected the NOx removal efficiency.
- (3) The self-made composite denitration agent used in real flue gas of ceramic industry can obtain a high NOx removal efficiency, which met the requirements of relevant national and local emission standard of ceramic industry.

Acknowledgments

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