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Pilot Study on Deep Denitrification from Municipal Solid Waste Incineration Flue Gas by Narrow Pulse Discharge Reaction Coupling with Wet Adsorption

Lingang WENG[a,b], Qinfeng SHI[b], Weiming LU[a], Keji QI[b,1], Qing YE[b], Anfei LUO[b], and Jinbiao WANG[a]
[a] Zhejiang DOWAY Advanced Technology Co., Ltd, Jinhua 321031, Zhejiang, China
[b] Zhejiang Engineering Research Center of Odor Control, Hangzhou 310000, Zhejiang, China

Abstract. To solve the issues existed in the traditional deep denitrification treatment technology for Municipal Solid Waste Incineration flue gas, such as complex system, large investment and high operating cost, a new method of deep denitrification by narrow pulse discharge reaction coupling with wet adsorption was proposed. The deep denitration of municipal solid waste incineration flue gas via the non-thermal plasma generated by the self-developed nanosecond pulse power corona discharge was studied. The results show that the removal efficiency of NO and NO_x was enhanced when the power supply frequency and residence time were increased. Under the case with single corona discharge reactor when the peak voltage is 82 kV, the frequency is 400 Hz and the residence time is 2s, the removal efficiency of NO and NO_x is 81.0% or 44.8% respectively. NO_x removal efficiency can be significantly improved by the process of narrow pulse discharge reaction coupling with wet absorption, the average concentration of NO_x at the outlet is 43.9 mg/m^3, and the average removal efficiency of NO_x is 64.1%, which is 19.8% higher than the efficiency of single narrow pulse discharge reaction, the narrow pulse discharge reactor has no obvious effect on the conversion of the original low concentration N_2O and CO in the municipal solid waste incineration flue gas, nor new N_2O and CO were produced. The research results of this paper have positive guiding significance for the industrial application of narrow pulse discharge reactor in the deep denitration of municipal solid waste incineration flue gas.

Keywords. Narrow pulse discharge reactor, denitrification, wet absorption, pilot test, municipal waste incineration

1. Introduction

Waste incineration has become the main means of domestic waste treatment in recent years because of its advantages of reduction and it possesses rapid growth in the proportion of domestic waste treatment [1]. Although waste incineration to some extent has solved the problem of large waste disposal area, the problem of flue gas pollution during waste incineration has caused widespread concern in society.

[1] Corresponding Author, Keji QI, Zhejiang Engineering Research Center of Odor Control, Hangzhou 310000, Zhejiang, China; Email: 1660972944@qq.com.

At present, environmental protection requirements are becoming increasingly stringent. Many local governments require that the NO_x emission concentration of flue gas from waste incineration plants be lower than 75 mg/Nm^3. In order to meet the emission requirements that may be further enhanced, the domestic waste incineration power plant puts forward the requirement of deep denitration, that is, the flue gas purification equipment should have the ability of NO_x emission level below 50 mg/Nm^3 [2]. Currently, certain domestic waste-to-energy plants are subjected to deep denitrification by using combined process technologies such as graded combustion [3, 4], SNCR in-furnace denitrification [5] and low-temperature SCR denitrification of the tail gas [6]. In order to meet the temperature requirements of low-temperature SCR denitration, flue gas heat exchange and reheat treatment are generally required [7]. The common flue gas heat exchange adopts gas-gas heat exchanger (GGH), and the flue gas reheat uses SGH (steam heating) [8], with large investment, high pressure drops and high operating costs. Narrow pulse discharge flue gas treatment technology has the ability of low temperature denitrification and has some engineering applications in flue gas denitrification of coal-fired power plants and steel plants [9, 10]. Its process system is simple, however, there are few researches in the deep denitrification of domestic waste incineration flue gas, and there is a lack of pilot studies for large flue gas volumes. In this paper, the self-developed nanosecond-level power supply and its matching bulk reactor are used as the core process to study the performance of nitrogen oxides removal in the flue gas at the back end of fabric filter, which provides relevant theoretical and practical basis for subsequent engineering promotion and application.

2. Test Device and Method

2.1. Test Equipment and Process Flow Introduction

The flue gas at the back end of the bag filter of a domestic waste incinerator in Zhejiang Province was selected for the pilot test of narrow pulse discharge reaction coupled with wet absorption deep denitrification, and the original purification process of the incineration flue gas was SNCR → semi-dry deacidification → dry deacidification → activated carbon adsorption → bag filter. This pilot test extracted 4000-16000 m^3/h flue gas at the back end of the bag filter for a deep denitrification pilot study. The system is shown in Figures 1 and 2, which mainly consists of narrow pulse discharge reactor, wet absorber and related control system, etc. The narrow pulse discharge reactor adopts needle-tube structure, and the power supply adopts the nanosecond high voltage pulse power supply of Zhejiang DOWAY. The waste incinerator flue gas was introduced to the narrow pulse discharge reactor system after SNCR furnace denitrification, semi-dry and dry de-acidification, activated carbon adsorption and bag filter. NO in the flue gas reacts with free radicals such as -O, -OH and -HO_2 generated by the narrow pulse discharge reactor and is converted into NO_2 and HNO_3, etc. After being absorbed and purified by the wet absorber, it was sent to the original main flue gas duct for discharge by the induced draft fan.

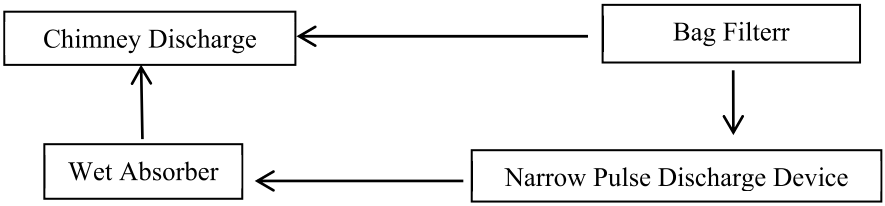

Figure 1. Process flow chart of this pilot project.

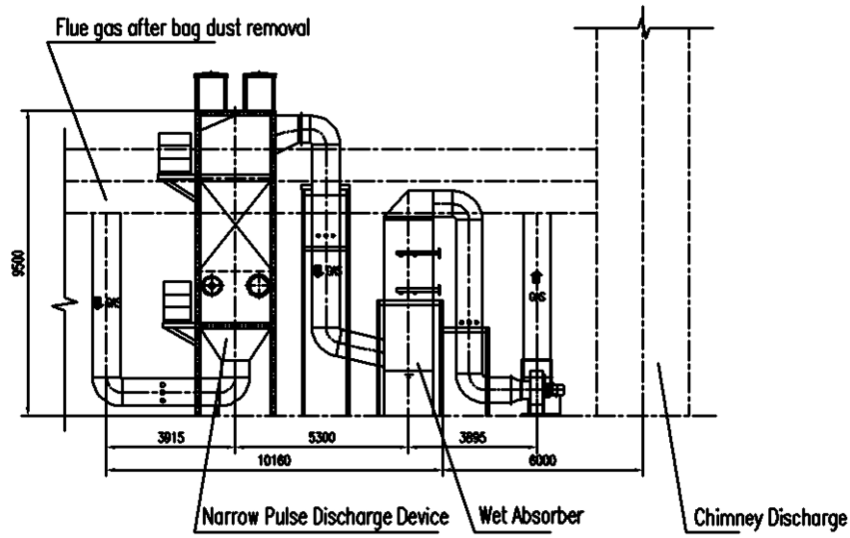

Figure 2. Side view of this pilot project.

2.2. Plasma Reactor and Power Supply Device

In this pilot test, the plasma reactor body adopts a pin-tube structure working under positive corona discharge mode. The peak voltage of nanosecond high-voltage pulse power supply was 100 kV, the peak current was 9 kA, the rising edge pulse width was less than 500 ns, and the working frequency was 100-500 Hz.

2.3. Test Flue Gas Condition

The gas for this pilot test was from the flue gas at the back end of the bag filter of a waste incineration plant in Zhejiang Province. By adding a variable frequency induced draft fan, the test flue gas flow was controlled to be 4000-16000 m^3/h, the temperature is 120-140°C, the O_2 content in the flue gas was 7.5%-11.2% (volume fraction), the water content was 16%-28% (volume fraction), the NO_x (basically all NO) concentration was 90-146 mg/Nm^3, and CO_2 content was 8%-10% (volume fraction).

2.4. Analysis and Test Methods

The flue gas parameters of this pilot project were measured by Wohler F550CI constant potential electrolysis method and Fourier infrared spectroscopy. The main measuring points were measuring point 1, measuring point 2 and measuring point 3 (Figure 2). The flue gas parameters at the entrance of narrow pulse discharge reactor were measured at point 1. The flue gas parameters after narrow pulse discharge reaction were measured at point 2. The flue gas parameters after plasma reaction coupled with wet absorption (Alkaline substances are NaOH) were measured at point 3, where NO_x is the concentration sum of NO and NO_2, calculated as NO_2.

The pulse voltage of the pulse power supply was measured by VD-150 UHV resistive capacitive voltage divider of North Star Company of USA, and the pulse current was measured by the 110 A+ ultra-wideband current sensor of Pearson Company of the United States, and the MSO/DPO5000B high-performance oscilloscope of TEK Company of the United States was used for display and calculation.

3. Results and Discussion

3.1. NO_x Removal Characteristics of Narrow Pulse Discharge Reactor

3.1.1. Effect of Flue Gas Residence Time on NO and NO_x Removal Efficiency

Residence time refers to the time required for flue gas to flow through the discharge area of the reactor, which is an important factor of narrow pulse reaction. In this pilot test, the flue gas flow was controlled by adjusting the frequency of newly added induced draft fan to achieve the purpose of the residence time adjustment. The residence time of flue gas in corona zone was 1s, 2s and 3s, the pulse peak voltage was 82 kV, the frequency was 400 Hz, and each residence time was tested for 50min to study the effect of the narrow pulse discharge reactor on the removal efficiency of NO and NO_x under different residence time conditions.

Figure 3 shows the effect of narrow pulse discharge reactor on NO and NO_x removal efficiency under different residence time.

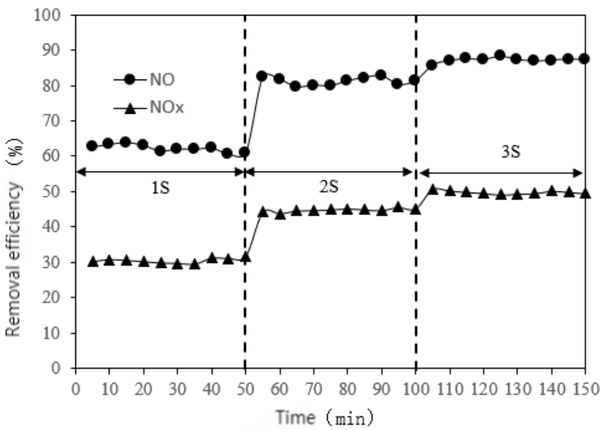

Figure 3. Effect of residence time on NO and NO_x removal efficiency.

In the process of narrow pulse discharge alone, the conversion of NO_x mainly depends on the active particles produced by corona discharge, such as -OH, -O, -O_3, -HO_2, etc. The main conversion reactions of NO_x are as follows [10, 11]:

$$NO + O \rightarrow NO_2 \tag{1}$$

$$NO + HO_2 + M \rightarrow HNO_3 + \mathrm{M} \tag{2}$$

$$NO + OH \rightarrow HNO_2 \tag{3}$$

$$HNO_2 + OH \rightarrow NO_2 + H_2O \tag{4}$$

$$NO_2 + OH \rightarrow HNO_3 \tag{5}$$

As can be seen from Figure 3, when the residence time was increased from 1s to 2s, the removal efficiency of NO and NO_x was significantly improved, from 62.1% and 30.5% to 81.0% and 44.8%, which were increased by 18.9% and 14.3% respectively. The reason is that with the increase of residence time, under the same discharge conditions and similar NO and NO_x concentrations, the reaction time of NO and NO_2 gas molecules and the active particles produced by pulsed corona discharge increases correspondingly, and the reaction is more sufficient. In addition, because the energy injected into the reactor per unit time is basically unchanged, the increase of residence time means the increase of energy consumption per unit flue gas, thus increasing the number of active particles per unit flue gas, so that NO and NO_2 react more fully with active particles, and the removal efficiency is higher. However, when the residence time increased from 2s to 3s, the removal efficiency of NO and NO_x increased to 6.1% and 5.0%, respectively, which was not obvious enough. Therefore, the optimal residence time under this pilot test condition is selected for 2s.

3.1.2. Effect of Frequency on NO and NO_x Removal Efficiency

The frequency of the pulsed power supply is an important factor. This pilot test investigated the effect of frequencies of 200 Hz, 300 Hz, 400 Hz and 500 Hz on the removal efficiency of NO and NO_x. The flue gas residence time was 2s, the peak pulse voltage was 85kV, and each frequency was tested for 50min to study the effect of narrow pulse discharge reactor on NO and NO_x removal efficiency under different pulse frequency conditions.

The results are shown in Figure 4, with the gradual increase of pulse discharge frequency from 200 Hz to 500 Hz, the NO and NO_x removal efficiency increased continuously, indicating that the increase of pulse discharge frequency was favorable for NO and NO_x removal, which is consistent with the results of Xu et al. [12] in their laboratory study. This is because as the pulse discharge frequency increases, the energy injected into the narrow pulse discharge reactor per unit time increases, and more gas molecules are excited, activated and dissociated to produce more active particles, facilitating the conversion of NO and NO_2. The rate of NO and NOx removal increased significantly with increasing frequency at first, and from 400 Hz onwards, the rate of increase tended to slow down, which is because with the further increase of frequency, although the number of discharges increases, the time of electron acceleration is shortened, and the rate of increase of the number of high-energy electrons that can excite gas molecules to produce active particles slows down. Therefore, when the frequency of pulse power supply is greater than 400 Hz, the removal efficiency of NO and NO_x will not increase obviously if the frequency is further increased.

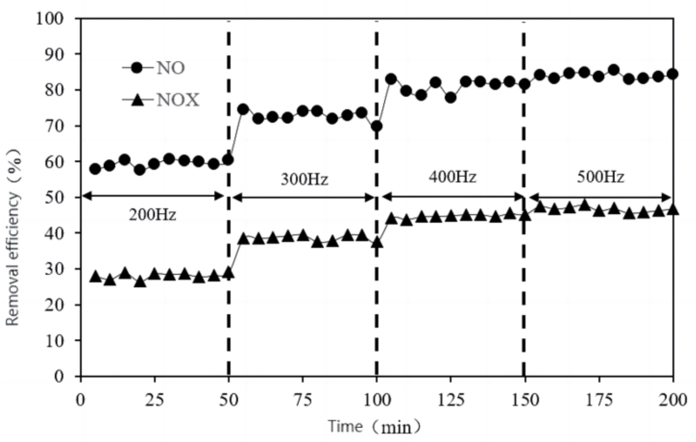

Figure 4. Effect of frequency on NO and NO_x removal efficiency.

3.2. Characteristics of NO_x Removal by Narrow Pulse Discharge Reaction Coupled Wet Absorption

The pilot test data of continuous operation from July 5 to August 10 were selected to analyze the effect of single narrow pulse discharge reaction (outlet concentration is measured at measurement point 2) and narrow pulse discharge reaction coupled with wet absorption (outlet concentration was measured at measurement point 3) on the removal of NO_x from flue gas.

Operating continuously from July 5th to August 10th, the residence time of flue gas in the narrow pulse discharge reaction device is 2s, the pulse power supply frequency is 400Hz, the pulse peak voltage is 80-92 kV (with the increase of moisture content in the flue gas, the peak voltage will increase accordingly). The wet absorption at the back end of the narrow pulse discharge reaction used NaOH absorption solution, and the circulating spraying liquid to gas ratio is controlled at 5 L/m^3.

The daily averages of NO_x for continuous operation are shown in Figure 5. The results show that under the purification effect of conventional process, the concentration of nitrogen oxides (more than 98% is NO) in the flue gas at the back end of bag filter could be controlled between 90-146 mg/Nm^3, and the average discharge/outlet concentration was 117 mg/Nm^3. The NO_x removal by single narrow pulse reaction was studied from July 5 to July 10 and from August 3 to August 12. The NO_x removal efficiency by single narrow pulse reaction was 42.3%-47.4%, with an average of 44.3%, and the average outlet NO concentration was 14.4 mg/Nm^3, and the NO conversion efficiency was 80.1%. The results from July 11 to August 3 show that the narrow pulse discharge reaction coupled with wet absorption process has a significant effect on NO_x removal, with an average outlet NO_x concentration of 43.9 mg/Nm^3 and a removal efficiency of 64.1%, which is 19.8% higher than the efficiency of single narrow pulse discharge reaction. In addition, the narrow pulse discharge reaction coupled with wet absorption promoted the absorption and removal of NO_2, as well as the co-absorption of 7.5% of NO. According to the energy consumption statistics during the continuous operation, the power consumption of the narrow pulse discharge reaction device was equivalent to 2.26 W· h/m^3.

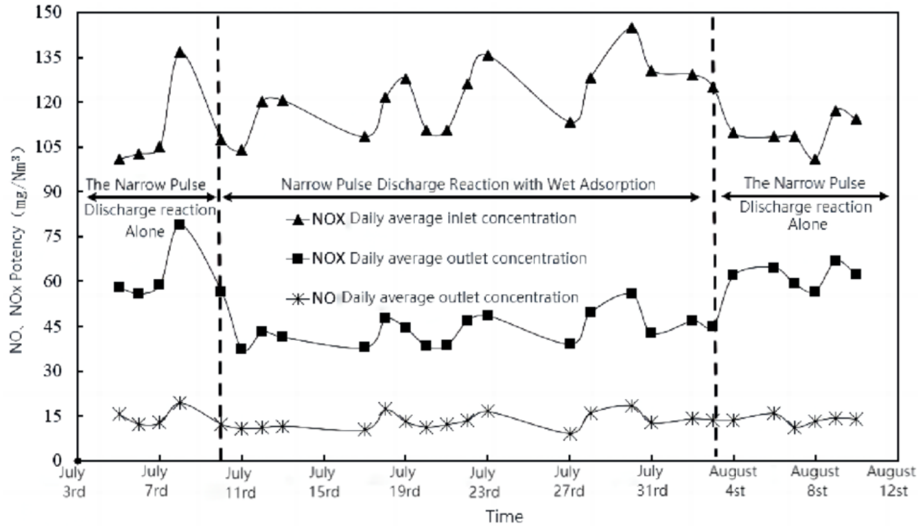

Figure 5. Variation of NO and NO_x concentrations at different measurement points.

The main chemical reactions are as follows [13, 14]:

$$NaOH + HNO_3 \rightarrow NaNO_3 + H_2O \quad (6)$$

$$2NaOH + 2NO_2 \rightarrow NaNO_3 + NaNO_2 + H_2O \quad (7)$$

$$2NaOH + NO_2 + NO \rightarrow 2NaNO_2 + H_2O \quad (8)$$

$$SO_3^{2-} + 2NO_2 \rightarrow 2NO_2^- \quad (9)$$

The water quality analysis shows that the nitrate (N molar ratio) in the solution accounts for about 78.5% and nitrite only accounts for 21.5%. This is mainly because of the high water content in the waste incineration flue gas, with a concentration of 16%-28%. The proportion of -OH radicals in the active particles generated by corona discharge is higher, and more HNO_3 is generated from NO after a series of reactions, which is more converted into nitrate after being absorbed by the alkali solution.

The wastewater generated by wet absorption can be remitted to the wastewater zero discharge treatment system after preliminary treatment, part of the NO_x is converted into N_2 by biological denitrification, and the rest can be concentrated and then evaporated and crystallized back into the flue gas of the spraying furnace to be captured by the bag filter and then entered the fly ash treatment system with the fly ash.

3.3. Effect of Narrow Pulse Discharge Reactor on N_2O, O_2 and CO in Flue Gas

In this pilot test, the variation of N_2O, O_2 and CO in the flue gas were investigated and studied when the narrow pulse discharge reactor was started and stopped. The narrow pulse discharge reactor was stopped during the time at 19:30-20:00 and 22:15-22:30, and at other times the reactor was put into operation. The results are shown in Figure 6.

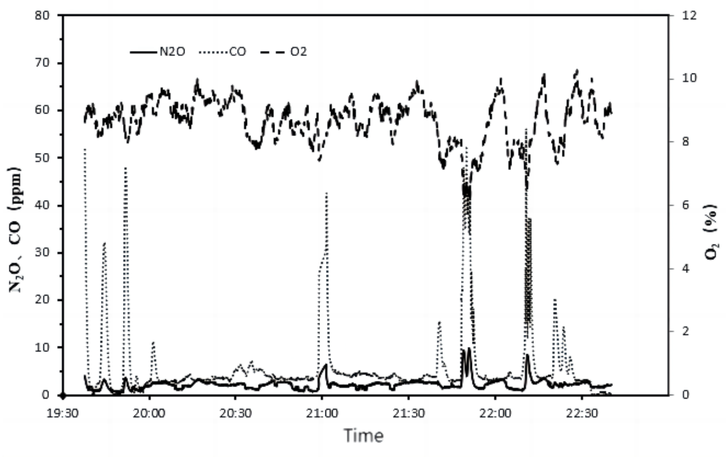

Figure 6. Plot of the variation of N_2O, O_2 and CO in flue gas.

The results show that the average concentration of original N_2O in flue gas was 1.44 ppm during the closing period of narrow pulse reactor from 19:30 to 20:00, 2.41 ppm during the closing period of narrow pulse reactor from 22:15 to 22:30, and 2.28 ppm during the opening period of narrow pulse reactor. It can be seen that the difference in N_2O concentration at the reactor outlet between the open and closed states of the narrow pulse discharge reactor is not significant, so the effect of the narrow pulse discharge reactor on the conversion of N_2O is not obvious, and the same conclusion is reached for CO. During the test period, there was a sudden increase in CO and N_2O, and this trend had the same variation as the sudden decrease in O_2 in the flue gas, which was judged to be an accidental phenomenon of boiler combustion, resulting in a rapid decrease in O_2 concentration and a sudden increase in N_2O and CO concentrations in a short period of time.

4. Conclusions

The removal efficiency of both NO and NO_x by the single narrow pulse discharge reactor increased with increasing power supply frequency, however, the rate of increase became increasingly slow. The increase of flue gas residence time in the pulsed discharge reactor significantly affects the NO and NO_x removal efficiency. When the peak voltage was 82 kV, the frequency was 400 Hz, and the residence time was 2s, the removal efficiency of NO and NO_x in the single narrow pulse discharge reactor was 81.0% and 44.8%, respectively.

The narrow pulse discharge reaction coupling with wet absorption process has a more obvious effect on NO_x removal and can achieve better removal efficiency of NO_x, with average outlet NO_x concentration of 43.9 mg/Nm^3 and average NO_x removal efficiency of 64.1%, which is 19.8% higher than the efficiency of the single narrow pulse discharge reaction.

The narrow pulse discharge reactor has no obvious effect on the conversion of the original N_2O and CO in the domestic waste incineration flue gas, and basically will not produce new N_2O and CO.

It was proved by the pilot run for a long time that the flue gas of domestic waste incineration was treated by narrow pulse discharge reaction coupled with wet absorption process on the basis of conventional purification process, and the outlet NO_x concentration of the exhaust flue gas was less than 50 mg/Nm^3, and the energy consumption of the narrow pulse discharge reaction device was about 2.26 W· h/m^3. This proposed process has high efficiency and stable performance for NO_x removal, which is worthy of engineering promotion.

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