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Removal Rate, Energy Yield and Energy Cost of NOx Removal by Corona Plasma

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Abstract. Pulsed corona discharge for NO_X/SO_2 destruction has been studied for many years, but how to achieve higher removal rate and energy yield of pollutants remains a critical issue. This paper studied enhancement of the energy yield of NOx removal by additive activated with corona dicharge. The experimental results demonstrate that corona radical injection can decrease the energy consumption, increase the removal rate and the energy yield of NO_X removal. When ammonia and propene are injected from nozzle electrodes supplied with pulse power supply, NO and NO_x removal rate can be increased by about 8% and 10% at a lower consumption compared with additives injected from the inlet of reactor.

Keywords: Corona plasma, Additive, Hydroxyl radical, Energy yield, NO_x

1. Introduction

Rapid development of world economy consumes more and more fossil fuels and emits more and more gaseous, aqueous, and solid wastes, which poses significant threats to water, atmosphere and soil environments. During combustion of fossil fuels including coal, gasoline, etc, gaseous pollutants including sulfur dioxide, nitrogen oxides, and carbon monoxide would be released into air. Commonly, nitric and sulfur oxides in the air would finally deposit on the land. The acidic dry or wet deposition of nitric and sulfur oxides harms the ecosystems and corrodes buildings, etc. Besides, through various means, the emission of these pollutants can cause harm to human health. So control of these pollutants' emission is critical to keep the health of ecosystem and humans.

In recent years, pulsed corona plasma for purifying flue gas containing toxic gaseous pollutants have been under intensive investigations [1-8], and these studies demonstrated the effectiveness of plasma technologies in destructing volatile organic compounds, sulfur dioxide, and nitric oxide. Early in the 1980's, Dinelli et al had conducted industrial-scale experiments in a coal thermal power plant [5]. Later, several larger scale industrial experiments were conducted in Korea and China [9-11]. These industrial experiments confirmed the physical feasibility of corona discharge process for simultaneous removal sulfur dioxide and nitric oxide. Owing to its big treatment capacity and simple structure, wire-to-plate type electrode structure had always been utilized as discharge electrode in the industrial application researches [9-11].

Although pulsed corona discharge for SO_2 and NO_x removal has shown its promising application prospect, some problems still exist. Including realization of the reliable and durable large-power pulse

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supply, how to get higher removal rate of pollutants at lower energy consumption remains one of the crucial issues. In this paper, we combine corona discharge radical shower (CDRS) technology and pulsed corona discharge (PCD) for NO_X removal, experiments of nozzle electrodes for additives injection supplied with pulse power supply for NO_x removal have been conducted. As far as we know, this novel process first uses CDRS for propene and ammonia injection to increase NO_x removal of PCD. Our object is using the corona radical injection technology improves the utilization efficiency of additives and generates more radicals responsible for NO_x removal, and then enhances the pulsed corona discharge NO_x removal and increases the energy yield of NO_x removal.

2. Experimental Method

2.1. Sampling and Testing

In experiments, synthesized gas with nitric oxide or sulfur dioxide and clean air was used as simulated flue gas. The flow rate, the humidity and the temperature of flue gas were measured by Hygropalm gas parameter testing apparatus (Rotronic, Swiss); the concentrations of nitrogen oxide was monitored online by NOA-305A portable Analyzer (Shimadzu, Japan) . If not specified, the flow rate of flue gas was fixed at about 170 m^3/h; the temperature in plasma reactor was controlled in the range of 60 ℃-80 ℃; the typical concentrations of nitric oxide, nitrogen oxide in flue gas were controlled at about 50-160 ppm, respectively. The humidity of flue gas was regulated through injection of water vapor into flue gas.

The pulse high-voltage is supplied by a positive narrow pulse power supply with a rotary spark gap switching. The capacitance of the tank capacitor is 100nF, and the capacitance of pulse-forming capacitor is selected in the range of 0.8~2nF. The voltage and current signals were sampled and recorded by a digital oscilloscope (HP54810A) with voltage probe (EP-100K) and current probe (Tektronix A6302) respectively. The discharge energy (E_P) is calculated by digital oscilloscope according to formula (1), the electric discharge power (P) is obtained by formula (2), and the specific input power (SIE) is calculated with formula (3).

$$E_p = \int_0^t VIdt \tag{1}$$

$$P = fE_p \tag{2}$$

$$SIE = P/Q \tag{3}$$

$$EY = \frac{\Delta[C]}{SIE} \times M \times 0.161\,(g/kWh) \tag{4}$$

$$EC = \frac{SIE}{\Delta[C]} \times 229\,(eV/molecule) \tag{5}$$

The energy yield (EY) and the energy cost (EC) are calculated by formulae (4) and (5), respectively, where the numbers of 229 and 0.161 are the conversion factor at 273 K, 1atm, the Δ[C], SIE and M represent the removal amount of NO or NOx in parts per million, the specific input energy and the molecular weight. The unit of SIE is kJ Nm^{-3}.

2.2. Configuration of Plasma Reactor

The nozzle-pipe-plate type corona radical injection system consists of six nozzle electrodes and two stainless steel plates. Six nozzle electrodes, each nozzle electrode made up of a stainless steel pipe with outer diameter of 10 mm and 20 nozzles with inner diameter of 1 mm, were separated into two groups for C_3H_6 and NH_3 injection, respectively. The spacing of adjacent nozzles (d) and the length of nozzles are 50mm and 20 mm, respectively. The nozzles are parallel to the stainless steel plates. The pulsed corona discharge system, consisted of 22 rectangular corona wires (4×4mm) and two stainless steel plates The wire-to-plate spacing and the wire-to-wire spacing are 75 mm and 90mm respectively. The cross-sectional area and the volume of the corona discharge reactor are 0.15 m^2 and 0.675 m^3.

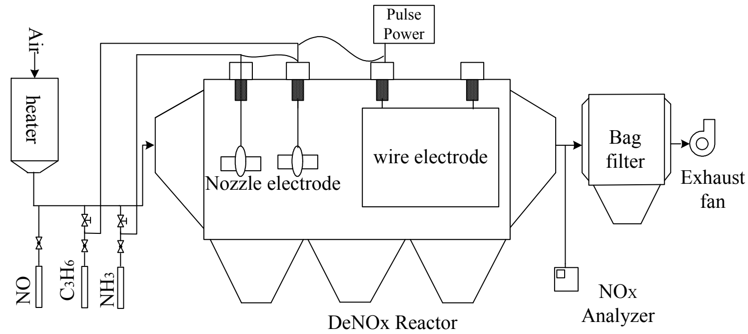

Fig.1: Configuration of corona discharge reactor

3. Results and Discussion

3.1. Effect of PCRI on NOx Removal

Fig. 2 and 3 show the removal rate, the energy cost and the energy yield of NO&NO_X at different injection ways of additives. The initial concentration of NO, the flow rate of simulated flue gas, and the specific input energy are 105 ppm, 171m^3 h^{-1}, and 1.5Wh m^{-3}, respectively. Ammonia and propene are injected into the reactor according to the molecule ratio of NO as 1:1. Compared with C_3H_6 and ammonia injected into flue gas from the inlet of reactor, higher removal rates of NO and NO_X can be gained at a slightly lower energy cost when C_3H_6 and NH_3 injected from nozzles (PCRI). In this experiment, the removal rates of NO and NO_X are increased by 7% and 10%, respectively.

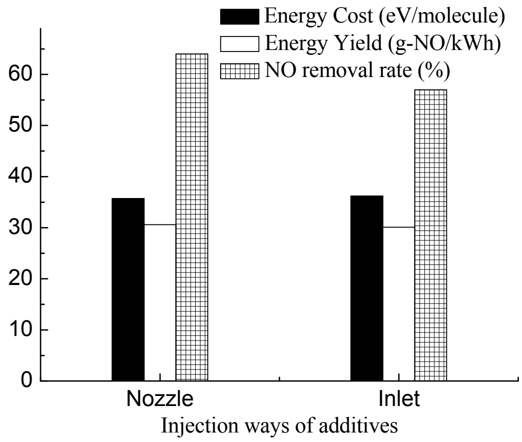

Fig.2: Effect of injection ways of additives on NO_X removal

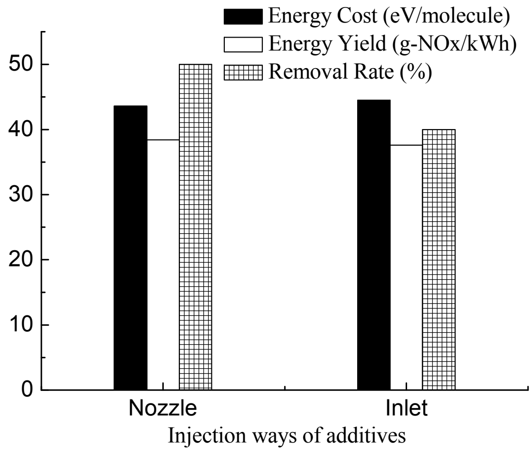

Fig.3: Effect of injection ways of additives on NO_X removal

The reason of PCRI enhancing NO_x removal can be explained as follows: when additives such as NH_3 and C_3H_6 injected from the corona discharge nozzles, the additives can collide with more radicals, electrons, and other active species because they must pass the corona zone between the nozzles and the grounded plates where abundant active species (radicals, electrons, ions, etc) exist. So more species such as NH, NH_2, RO_2 and HO_2, etc responsible for NO removal can be produced.

3.2. Effect of C_3H_6 on NOx Removal

Hydrocarbon has been demonstrated as an effective additives for lowering energy consumption of NO_X removal [16-18], and though no harmful byproducts such as HCN are detected，however, small amounts of CH_3COOH were detected when C_2H_4 used as additives [16], and few C_2H_6 & HCHO were detected when C_3H_6 infused into flue gas [17]. Though these undesirable byproducts such as aldehyde and ketone can also be degraded by pulsed corona discharge [18], an appropriate input amount of C_3H_6 should be selected.

Fig.4 and 5 show NO and NO_X removal as a function of C_3H_6 input. Unlike the NO_X removal, NO removal increases sharply with C_3H_6 input. In this experiment, NO removal rate reaches over 90%, comparing to the NOx removal of about 27%. The reason can be explained as follows：NO is removed mainly through conversion to NO_2 when C_3H_6 is injected into flue gas, so NO_2 increases sharply with C_3H_6 input, but NO_X changes slightly. When NO is treated only with pulsed corona discharge, produced NO_2 is about 50% of converted NO, but the value rises over 75% when C_3H_6 is injected. This phenomenon has great difference to that when NH_3 is injected.

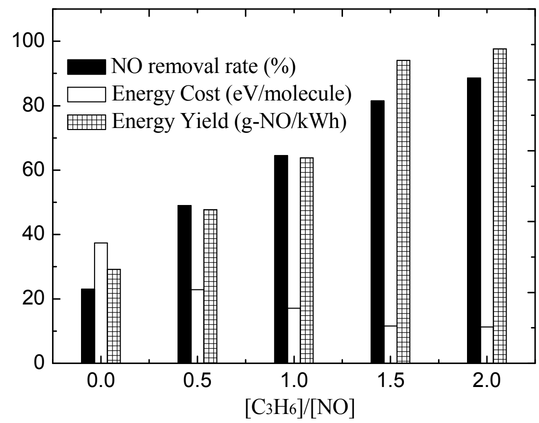

Fig.4: Effect of injection ways of additives on NO_X removal

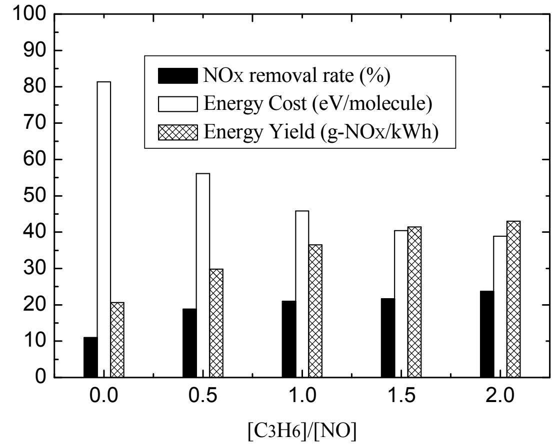

Fig. 5: Effect of injection ways of additives on NO_X removal

3.3. Effect of Initial Concentration of NO on NOx Removal Rate

Effect of initial concentration of NO on NO/NO_X removal is shown in Fig.6 and 7. NO/NO_X removal increases with specific input energy and decreases with initial concentration of NO. At energy consumption of 8.6kJ Nm^{-3}, NO and NO_X removal efficiency can reach 76% and 60% when initial concentration of NO being 55ppm.

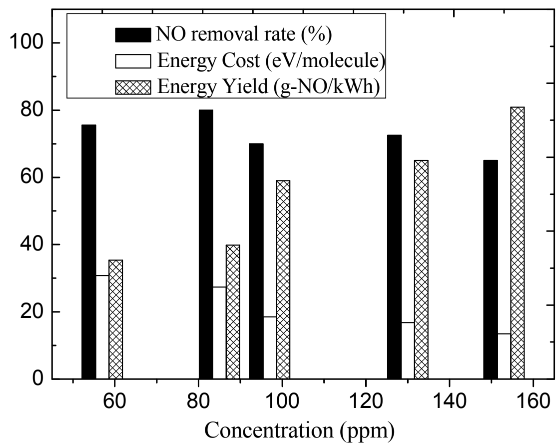

Fig.6: NO removal under different initial concentration

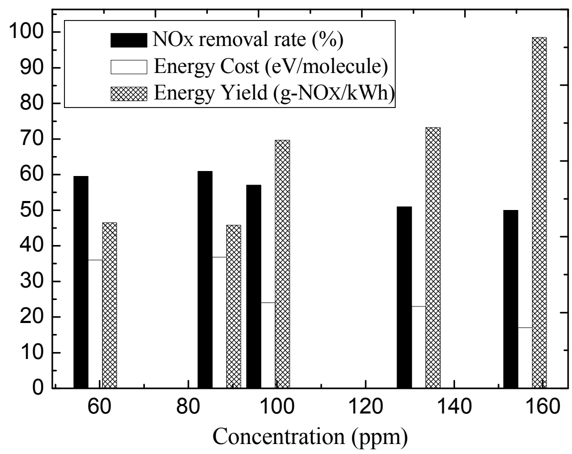

Fig.7: NOx removal under different initial concentration

4. Conclusions

Experimental results show that additive and corona radical injection play a great role in NO removal, but for effective NO_X removal, additives responsible for production of reductive radicals or capable of easily reacting with NO_X are required. Additive (e.g. C_3H_6) and corona radical injection had important influence on removal rate, energy cost and energy yield of NO/NOx removal. And higher energy yield of NO/NOx removal was achieved under lower initial concentration of NO and higher injection quantity of C_3H_6.

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