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Progress in Study of NOx Removal from Flue Gas by Non-thermal Plasma

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ABSTRACT

Tests for removal of nitrous oxides are performed by simulating flue gas using self-designed and self-developed plasma generator in this experiment; research and analysis show that there is a direct and linear relationship between the efficiency of denitration and the input power of plasma at normal temperature. The plasma generator begins to discharge at an input power of 15w to remove the nitrous oxides; the removal efficiency improves continually with the increase of plasma input power; then, it maintains the efficiency of over 80% when the input power is 35w, displaying a weak relation between the two factors. From the aspects of technology and economics, removal efficiency can be improved by high input power, but it is of huge energy consumption, being not economically feasible. Meanwhile, brief experimental analysis on removal efficiency is performed by adding sulphur dioxide into the mixed gas.

INTRODUCTION

Nowadays, the main source of energy comes from the process of coal burning, in which thermal power is transferred to other forms of energy, constituting the major way of utilization. Meanwhile, large amount of SO_2 and NOx are released, being the sources of smog and acid rain and the killer of air quality, thus severely pollute the environment and pose threat to human health (Gao et al. 2013). It is becoming a major strategic issue on sustainable development to control the SO_2 and NOx in waste gas effectively and economically. The tighter global standard of pollution discharge encourages researchers of every nation to actively develop new technology and to find new ways on denitration and desulfurization (Wei et al. 2001). Presently, researches from home and abroad have achieved progressive breakthroughs, developing numerous advanced technologies, such as, desulfurization by coal washing, denitration and desulfurization inside stoves and of flue gas. Low-temperature plasma technology being the most popular new way in control of the SO_2 and NOx discharge, the authors made extensive explorations on denitration and desulfurization by aid of a variety of electrode generator systems such as wire-tube model, wire-plank model and needle-plank model. There is still distance between these studies and actual application due to the complexities of actual reaction and other factors (Zhao et al. 2007a). Especially, this is significant in terms of the process and condition of denitration and desulfurization at the same time, such as temperature, water, flow velocity, effect of oxidation of SO_2 and NH_3 on denitration, the consumption of additive NH_3 and its secondary pollution which requires further study (Zhao et al. 2007b). This paper focuses on the efficiency of desulfurization in self-designed plasma generator under certain conditions and on the comparison of denitration efficiency between denitration alone and with desulfurization combined. This experimental analysis on the relationship between energy consumption of plasma generator and denitration efficiency aims to search for new methods of minimizing energy consumption and to industrial application.

MATERIALS AND METHODS

Plasma Generator

Stainless steel is applied to the entire body of the generator. The discharging area consists of 150mm of length and 3mm of space (single-sided). Medium resists the discharging process where, the smaller the space, the lower the input and discharge voltage. The thickness of resistance medium also affects the discharge of electrodes to some extent. Steel pipe: 25*20, gear-shape electrode core: 10mm, diameter of raised stand : 14mm. Structure of the generator is shown in Fig. 1.

Experiment Platform

Gas supply system: The simulated gas in the experiment consists of N_2, O_2, NO, NH_3 and SO_2. Since NO in coal-generated electricity plant composes 95% of the NOx, NO_2 accounts for only 5%, which is relatively low and can be neglected. Therefore, the experiment introduces standard NO gas to simulate NOx in the gases from electricity plant (Svachula et al. 1993, Liu et al. 2004). Velocity of gas is controlled by rotameter from each steel tube, amalgamated in the mixer and the well-mixed gases are to be led into gas heating tube. To avoid oxidation of NH_3 during heating process, the gas is infused separately from the heating tube into the reactor. The system is shown in Fig. 2.

Gas analysis system: Application of the online gas analysis equipment MGA-5 from German MRU allows online measurement of the density of NO, NO_2, SO_2, NH_3, CO and O_2, achieving the accuracy of ±1ppm. The inserted gas sampling pump extracts sample gas from the flue through the sampling pipe and tube. When the sampled gas is dehydrated and de-sooted by gas-water filter, the inserted electrochemical sensor tells the density of every component gas.

Reaction Mechanism (Yan et al. 2005)

$O_2+e_* \rightarrow 2O+e$

$H_2O+e_* \rightarrow H+OH+e$

$O_2+O \rightarrow O_3$

$H+O_2 \rightarrow HO_2$

$NO+O \rightarrow NO_2$

$NO+O_3 \rightarrow NO_2+O_2$

$NO+HO_2 \rightarrow HNO_3$

$NO_2+OH \rightarrow HNO_3$

The research conducted by Young Sun Mok (1998) showed that the most important active group is ozone in the process of transforming NO into NO_2, while the active groups -O, OH, HO_2, N- are sources of ozone. Besides the experiment, many researchers did the mathematical simulation of plasma reactions of SO_2 and NO_x. By solving the drift and diffusion currency equation and Poisson equation, Kulikovsky (1997) found the electronic concentration distribution in discharge lingers and the concentration of active group resulted from collision ionization. Then, he determined active group and chemical kinetics reaction and finally reached a conclusion that O_3 and OH are the main active species besides SO_2 and NO_x.

Decided by reaction mechanism, the experiment produces a certain concentration of NO_2 while removing NO. But NO_2 is easy to deal with, so it was not calculated in the removal efficiency of NO_x.

EXPERIMENTAL RESULTS AND DISCUSSION

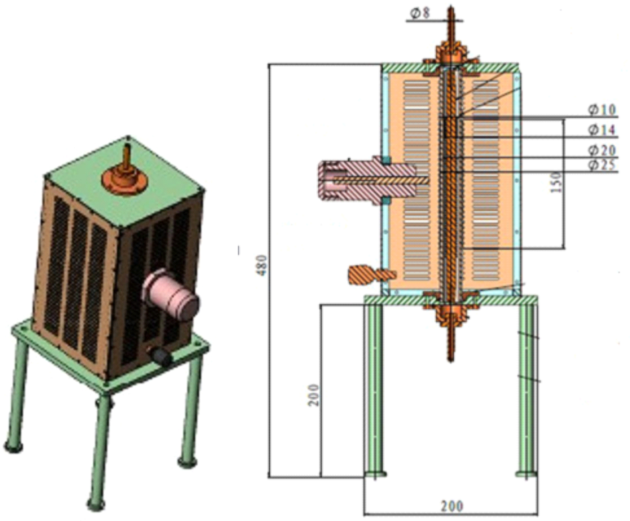

Fig. 1: Structure of the generator.

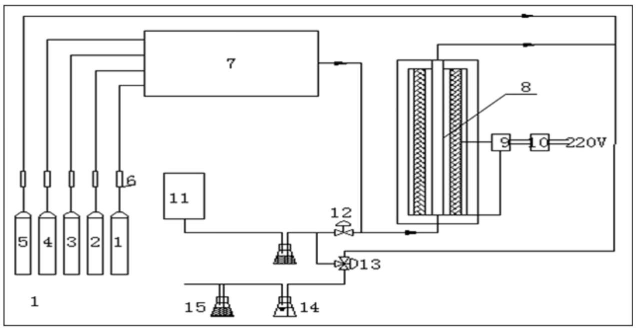

Fig. 2: Process of test platform.
1-O_2 bomb, 2-SO_2 bomb, 3-N_2 bomb, 4-NO bomb, 5-NH_3 bomb, 6-flow counter, 7-gas mixer, 8-plasma generator, 9-plasma electric source, 10-voltage transformer, 11-gas analysis equipment, 12-valve, 13-three-way valve, 14-inverted bottle, 15-dry bottle

Analysis of NOx removal from flue gas by non-thermal plasma: The NOx removal efficiency of low temperature plasma reactor under different levels of power is shown in Fig. 3.

The removal efficiency of catalyst improves continually with the increase of plasma input power. The removal efficiency is over 80% when the input power is over 35W and it reaches 91.7% when the input power is 52.8W. The removal efficiency is in positive proportion to input voltage and current. However, the increase is not prominent. When the voltage reaches a certain high level and dielectric barrier discharge is punctured, the removal efficiency will decrease sharply.

Denitration efficiency analysis under simulated flue gas flow increase: The denitration efficiency on the increase in simulated flue gas flow conditions is shown in

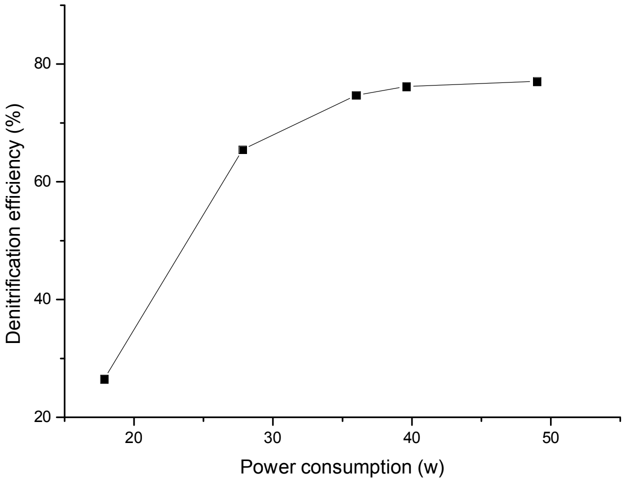

Fig. 3: Analysis of NOx removal from flue gas by non-thermal plasma.

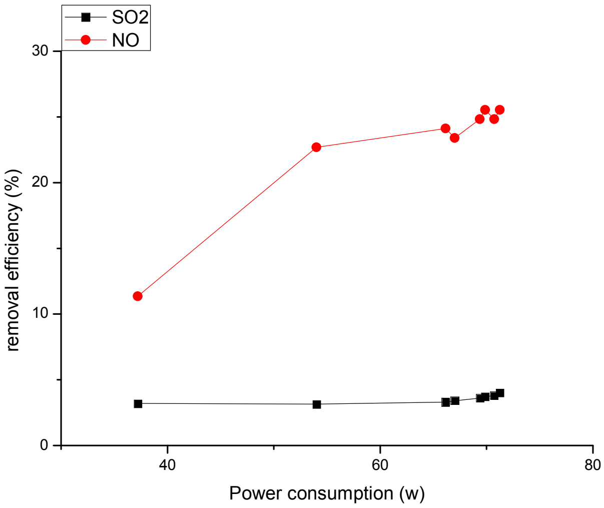

Fig. 5: Analysis of the removal efficiency of simultaneous desulfurization and denitration.

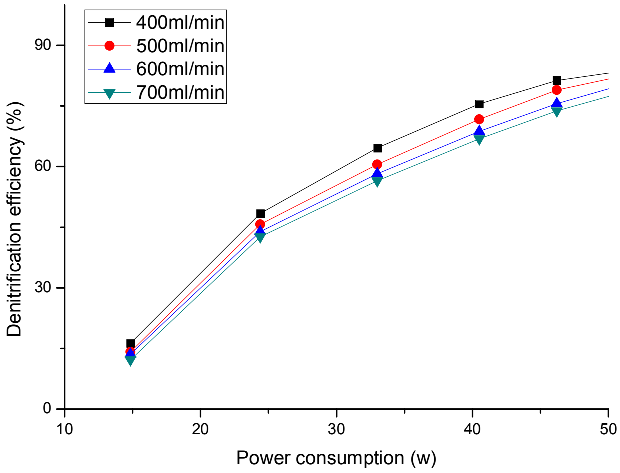

Fig. 4: Analysis of denitrification performance of variable flow.

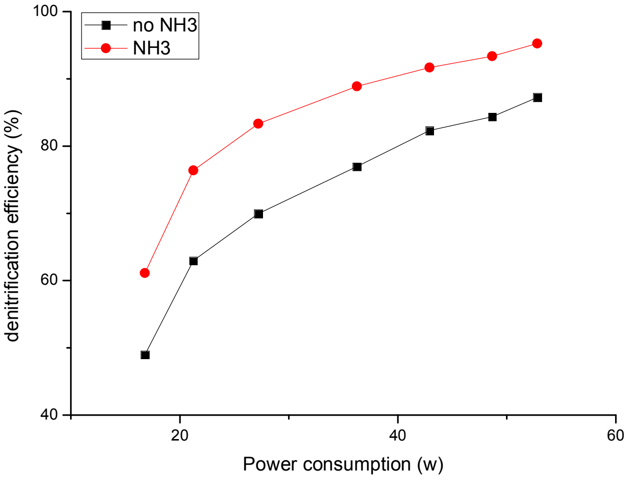

Fig. 6: Ammonia and non ammonia denitration performance comparison.

Fig. 4

As the flue gas flow increased, compared with a small flow of flue gas denitration efficiency, decreased the efficiency. Power consumption under the same plasma reactor conditions, about denitration efficiency decreased by 10%, and the reaction mechanism of the plasma is consistent, open more molecular bond, need to consume more energy, energy is conserved. In the same input voltage and current situation, four kinds of different flow simulation of flue gas denitration efficiency, as shown in Fig. 4, the greater the flow, the denitration efficiency compared to smaller, minimum flow, denitration efficiency value in the curve on the top.

Analysis of simultaneous desulfurization and denitration: The experimental results of the removal efficiency of simultaneous desulfurization and denitrification is shown in Fig. 5.

The addition of SO_2 influences the removal efficiency quite obviously. When the input voltage is increased to 40V, the removal efficiency still lingers under 80%. It is almost of no use in removal of SO_2 because the concentration statistics change little through the experiment, but it is of great importance in the removal of NO. By studying simultaneous desulfurization and denitration of gas flow using wire-plate reactors, Institute of Electrostatics of Dalian University of Technology reached the results as follows (Zhu et al. 1998):

1. The addition of NH_3 accelerates the oxidation of SO_2. In total removal rate, the oxidation of SO_2 is less than 30% in gaseous phase and aerosol surface reaction is over 50%, while thermo-chemical reaction takes up only 20%.
2. Moisture content in gas flow greatly influences the removal, energy utilization, and products.
3. NO is oxidized into NO_2 by O and O_3 which results from oxygen discharge. NO_2 is removed by free radical. The addition of H_2O is more important than of O_2 whereas, the synthetic effect of H_2O and O_2 is better in removing NOx than that of either.

Effect of NH_3 on denitrification performance: Ammonia added a certain improvement on denitration efficiency, under the same conditions, and the denitration efficiency Qu Xianru diagram without ammonia is shown in Fig. 6.

The addition of ammonia, in high energy electron effect, make the NH_3 decomposed into active particles, and NO reaction is more active, so the denitration efficiency further improved.

CONCLUSION

According to the analysis of experimental research, the conclusions of NOx removal from flue gas by non-thermal plasma are as follows:

1. The removal efficiency is over 80% when the input power is over 35W. The removal efficiency of catalyst improves continually with the increase of plasma input power.
2. Under the same power consumption of plasma reactor, the efficiency decreases. This coincides with the reaction mechanism of plasma. The more molecular bonds opened, the more power will be consumed according to energy conservation.
3. The addition of SO_2 influences the removal efficiency significantly. It is almost of no use in removal of SO_2, but it is of great importance in that of NO. Some document analysis (Liu et al.2006) state that it is because SO_2 digests OH, that the NO-reaction-available OH decreases.

A conclusion can thus be safely drawn that the technology of NOx removal by non-thermal plasma is still in the experimental stage. It requires more efforts of researchers, reducing energy consumption, enhancing the removal efficiency so as to achieve higher industrialization.

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