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Characteristics of NO reduction with non-thermal plasma

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Abstract: As a new type of NO removal system, NO reduction in N_2-NO plasma was applied to solve the difficulties in the traditional methods, such as higher energy-consumption, larger equipment size and high cost, and so on. Using the experimental NO reduction system with single-pair electrode tip discharge structure, the NO reduction characteristics of N_2-NO system were revealed to guide the engineering practice; the results of NO reduction with single-pair electrode tip discharge plasma also have the same instructive meaning to the NO reduction with multi-pair electrode tip discharge plasma. The amount of both active N atom and NO removal rate increased with the distance l_g increasing between the two electrode tips and then dropped when the distance exceeded a certain value. The NO removal rate increased while the voltage between two electrode tips or the resident time of gas flow increased. The distance is a key geometrical variable factor that can determine the intensity of electric field between two electrode tips and the resident time of gas. In this paper, the effects of the dielectric features on NO reduction using dielectric-barrier discharge plasma system were also studied. The results of NO removal rate with different dielectrics such as Al_2O_3, CaO, MgO and glass showed that the electric field intensity is different with different dielectric, because it brings different energy to particles in discharge room and thus it causes different NO removal rate.

Keywords: NO reduction; plasma; characteristic; single-pair electrode tip discharge; dielectric-barrier discharge

Introduction

Removing NOx with non-thermal plasma, following the conventional scrubber technologies such as dry way, wet way and half-dry way, has become a novel high-tech of denitration (Galkimberti, 1988; Vitello, 1994; Yamamoto, 2000). With many excellent characteristics of low investment, small operation spot, low operation expenses, etc, this technology has become an internationally accepted and new flue gas denitration technics (Shimizu, 1999; Orlandini, 2000; Rajanikanth, 2001; Kawasaki, 2001).

Several successful demonstration in power plants have been set up with the denitration technology of electron beam (EB) with high energy. Although pulsed corona induced plasma chemical process (PPCP), a more advanced technology than EB, is still under study in laboratory, many great achievements have been achieved in related areas like chemical reaction kinetics, gas phase electric discharge physics, ns grade high voltage pulse power and pulse corona reactor with the efforts of the scientists over the world. A new technology derived from it, combining plasma with adsorption, has also been studied extensively and made great progress in application. Although there are these fruitful achievements, many key theoretic and technological problems must be resolved before applying PPCP to denitration in power plant. Many researchers are still seeking new low-cost technology that can reduce reaction energy consumption and increase NO removal rate greatly.

NO reduction in N_2-NO plasma system has been proved by Zhang and Yu in laboratory to be an effective technology which can solve the difficulties in the traditional methods, such as higher energy-consumption, larger equipment size and more expensive cost, etc. (Zhang, 2002). This technology does not need very high voltage to break the N-O bond and the product of the system is innoxious gas mixture. Thus, this NO reduction technology also can be used to reduce NO in the exhaust gas emitted from the mobile engine. The characteristics of NO reduction in N_2-NO plasma system must be studied before it is applied to practice and to make an embedded and systemic research on the characteristics of the NO reduction in N_2-NO plasma system.

In this paper, single-pair electrode tip discharge (SETD) structure was emplied to study the effects of the physical feature of electric field and the electrode geometry on NO reduction. The voltage V_m between two electrode tips and the distance l_g between two electrode tips were used respectively to describe the effects of the physical feature of electric field and the geometric feature of electrodes. The characteristics of NO reduction in N_2-NO plasma system can be a guide to engineering practice and the results obtained here are also have the same instructive meaning to the NO reduction with dielectric-barrier discharge plasma. The effects of the dielectric features on NO reduction using dielectric-barrier discharge plasma system were also studied in this paper and the results of NO reduction rate with different dielectrics such as Al_2O_3, CaO, MgO and glass were obtained respectively.

1 Experments and mothods

1.1 Experiment to test the effects of V_m and l_g

1.1.1 Single-pair electrode tip discharge

The structure of single-pair electrode tip discharge (SETD) is showed in Fig.1 and the structure of multi-pair electrode tip discharge (METD) is showed in Fig.2.

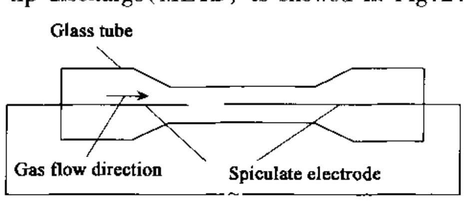

Fig.1 Schematic structure of SETD

Obviously, there is only a pair of tip electric poles in

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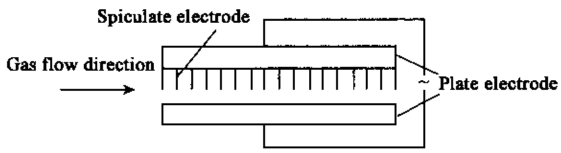

Fig.2 Schematic structure of METD

SETD, the gas flow direction is parallel to the discharge direction. In METD, many tip electrodes are opposite to a plate electrode and the gas flow direction is perpendicular to the discharge direction. The structure of SETD is simple and uniformity compared to that of METD and some variables could be adjusted easily by moving the two spiculate electrodes.

Besides the above advantages of SETD, the structure of SETD can eliminate the interference of the adjacent spiculate electrodes in the structure of METD. Thus, the structure of SETD is adopted in this experiment to study the effects of V_m and l_g on NO reduction and it can help us to investigate and master the essential characteristics of NO reduction more carefully.

1.1.2 Experimental setup

Fig.3 is the schematic experimental setup. The voltage output of voltage adjustor (model TDGC-2) is ranging between 0 and 240 V and its frequency output is 50 Hz. The maximum power output of the transformer (model HB708) is 50 W. Its highest voltage output and frequency output are about 15 kV and 30 kHz respectively. The gas analyzer (model SAE19, Germany) is used to measure the concentration of NOx, SO_2, and so on.

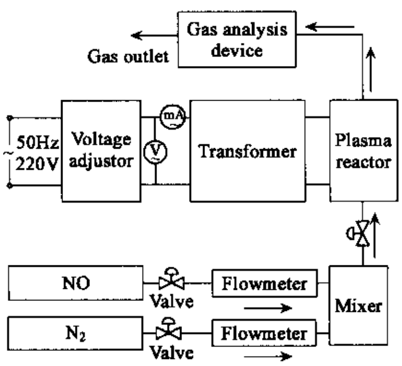

Fig.3 Schematic system for NO reduction experiment

1.1.3 Experimental methods

Gas mixture passing the plasma reactor is blended in a mixer by adjusting the flux of NO and N_2 respectively to have the needed initial NO concentration. The voltage between the two electrodes can be adjusted using the voltage adjustor. The distance between two electrodes can be adjusted by moving electrodes. When NO initial concentration, voltage and distance have been adjusted, the NO reduction rate can be measured using the gas analyzer at the outlet of plasma reactor.

1.2 Experiment to test the effect of dielectric

1.2.1 Dielectric-barrier discharge

The structure of dielectric-barrier discharge (DBD) is showed in Fig.4.

1.2.2 Experimental methods

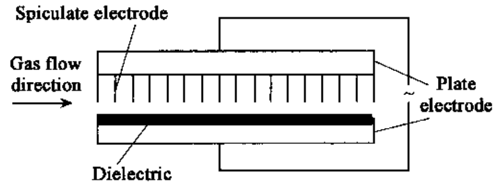

Fig.4 Schematic structure of dielectric-barrier discharge

One copperplate with tips and another copperplate without tips are placed in plasma reactor as electrodes. The copperplate without tips is covered with a dielectric and the dielectric can be changed among Al_2O_3, CaO, MgO and glass. The distance between the dielectric and the tips is 2 mm.

In the experiment, the voltage output of the voltage adjustor is fixed at 60 V. The highest output voltage of the transformer is about 10 kV and its frequency is about 30 kHz. The flux of NO and N_2 is fixed at 170 ml/min and 100 L/h respectively. Al_2O_3, CaO, MgO and glass are placed as the dielectric in experiment alternatively. The time of each set of experiment is 15 min.

2 Results and discussion

2.1 The NO reduction rate versus the distance l_g

Fig.5 shows the relation between NO reduction rate and the distance l_g between two electrode tips. It is quite clear that the NO reduction rate rises firstly and then falls with the increase of distance l_g. This is related with the discharge effect of single pair electrodes and reflects the contradiction between the diameter ϕ of discharge outline and the intensity E_g of the electric field.

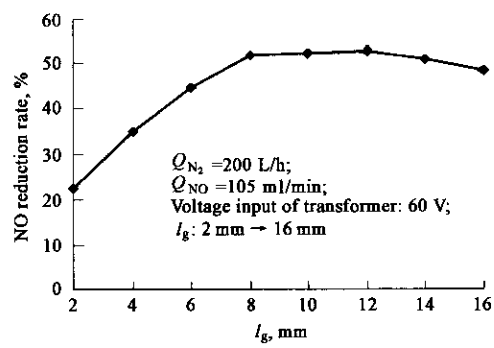

Fig.5 NO reduction rate versus the distance between two electrode tips

In the experiment, the discharge outline can be found to be very slim and its diameter is very small when the distance l_g between the two electrode tips is only 2 mm. When the distance l_g increases, the diameter ϕ of the discharge outline, the intensity of the discharge and NO reduction rate will all rise. According to the possible reaction of $e + N_2 \rightarrow e + N + N$ (Zhang, 2001), when the electron which get energy in the electric field passes the energy to N_2, there is redundant energy left. Consequently, only a proportion of N_2 can be collided by electrons and partial NO molecules can be reduced by the activated N atom because the diameter of the discharge outline is quite small. When the distance l_g increases, the enlarged diffusion extension of electrons will cause the increase of the diameter of microdischarge tunnel, therefore, the diameter of the discharge outline will go up and more N_2 and NO will join the reaction. So, the intensity of the discharge will be strengthened and the NO reduction rate will increase at the same time.

When the distance l_g is smaller than a certain value l_{gmax} (in this experiment, the value is 10 mm), the strengthening effect of the increasing diameter of the discharge outline will be obviously stronger than the weakening effect of the intensity of the electric field which is caused by the increasing distance l_g. When the distance l_g exceeds this value l_{gmax}, the discharge will begin to be weaken slowly with rising tip distance, and the NO reduction rate will decrease simultaneously. This indicates that the weakening effect of the electric field has surpassed the strengthening effect of the diameter of the discharge outline. From $E_g = \frac{V_m \varepsilon_d}{2 l_d \varepsilon_g + l_g \varepsilon_d}$ and $l_d = 0$, the expression of $E_g = \frac{V_m}{l_g}$ can be obtained. Here, l_d represents the thickness of the dielectric in dielectric-barrier discharge (DBD). ε_d and ε_g represent the dielectric constant of the dielectric and gas gap respectively. V_m represents the voltage between the two electrode tips. So, according to the expression $P_e = \frac{n_e e^2 E_g^2}{2 m_e} \times \frac{\nu_e}{(\omega^2 + \nu_e^2)}$, the total power P_e of the electron will go down with the distance l_g increase, and as a result, the NO reductive rate will decrease. Here, n_e is the concentration of electrons. m_e is the mass of the electron. ν_e is the collision frequency of electrons. ω is the oscillation frequency of plasma.

All the above can be explained as the following. When $l_g < l_{gmax}$, the energy which the electron gains will surpass the energy which is required to produce the active N atom in the reaction: $e + N_2 \rightarrow e + N + N$, and the redundant energy will be mainly used to ionize more N_2 which will come into the enlarged discharge outline when the tip distance increases, and as a result, the NO reduction rate is high. When $l_g = l_{gmax}$, the energy which is offered by the electric field is equal to the energy which is needed to ionize N_2 in the discharge outline. When $l_g > l_{gmax}$, the energy which the electron gains will be not enough to ionize the N_2 effectively, and this will cause the decrease of the NO reduction rate.

Therefore, for one certain structure of single-pair electrode tip discharge, there must be a best value l_{gmax} between two electrode tips and this value is the turn-point which distinguishes the main effect of either the diameter ϕ or the intensity E_g. This value can be determined by experiment. In the present structure of discharge, l_{gmax} is detemined to be 10 mm and the corresponding intensity E_g of the electric field is 10^6 V/m.

Because a couple of electrodes consume all the input power of the electrical source in the structure of single-pair electrode tip discharge and plural electrodes consume all the input power of the electrical source in the structure of the multi-pair electrode tip discharge, l_{gmax} of the structure of the single electrode tip discharge will not be the same with that of the structure of the multi-electrode tip discharge quantitatively.

2.2 NO reduction rate versus the voltage V_m

Fig.6 shows the relation between NO reduction rate and the voltage V_m between two electrode tips.

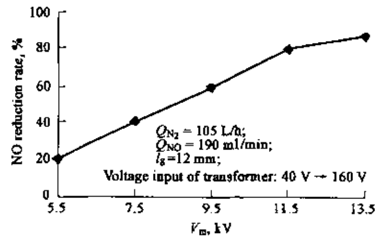

Fig.6 NO reduction rate versus the voltage between two electrode tips

It can bee seen that the NO reduction rate rises with the increase of V_m between two electrode tips. According to the expression of $E_g = \frac{V_m \varepsilon_d}{2 l_d \varepsilon_g + l_g \varepsilon_d}$, the intensity E_g of the electric field between two electrode tips goes up with the voltage increase, and the rise of intensity E_g of electric field causes the increase of the intensity of the single-pair electrode tip discharge and the power $P_e = \frac{n_e e^2 E_g^2}{2 m_e} \frac{\nu_e}{(\omega^2 + \nu_e^2)}$ obtained by the electron. As a result, in the reaction of $e + N_2 \rightarrow e + N + N$, more energy will be transferred to molecules of N_2 by electrons and more gas between two electrode tips will be ionized. More active radicals will be produced in this reaction and the NO reduction rate will be increased.

2.3 NO reduction rate versus resident time

Fig.7 shows the relation between NO reduction rate and the resident time of gas in the plasma reactor.

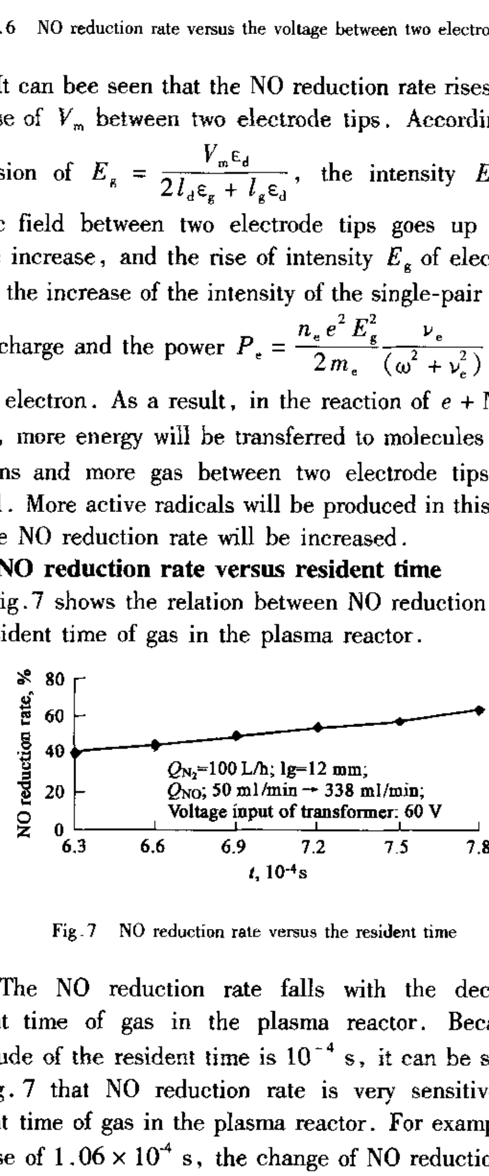

Fig.7 NO reduction rate versus the resident time

The NO reduction rate falls with the decrease of resident time of gas in the plasma reactor. Because the magnitude of the resident time is 10^{-4} s, it can be seen from the Fig. 7 that NO reduction rate is very sensitive to the resident time of gas in the plasma reactor. For example, in a decrease of 1.06×10^{-4} s, the change of NO reduction rate is about 15.97%.

In plasma reactor, all active radicals will move to the outlet of the reactor along with the gas flow. There are part of radicals will not collide with the molecules of NO and N_2 if the resident time is small. As a result, the NO reduction rate will be decreased.

2.4 Distance l_g

It can be seen from the above, the distance l_g is a geometrical variable. l_g is one of the factors which determine the intensity E_g of the electric field between two electrode tips and the resident time of gas in the plasma reactor and determines the NO reduction rate.

Firstly, the distance l_g is related to the intensity E_g of the electric field between two electrode tips and the diameter ϕ of discharge outline. Secondly, it is also related to the resident time of gas in the plasma reactor. Therefore, in practical operation, the best distance l_{gmax} must be obtained firstly through experiments under a fixed flux of gas and l_g will be fixed to the best distance l_{gmax} before using appropriate flux.

In the single-pair electrode tip discharge, because the airflow direction is parallel to the placed direction of two electrode tips, the above method can be used. But in the structure of multi-pair electrode tip discharge such as dielectric-barrier discharge, because the gas flow direction is perpendicular to the placed direction of electrode tips, more electrode tips can be added to increase the length of discharge field so that the resident time of gas can be increased.

2.5 Effect of dielectric

Table 1 shows the effect of dielectrics on NO reduction.

Table 1 NO reduction rates with 4 different dielectrics

Dielectric material	Dielectric constant, ε_d	Concentration of NO before reaction, ppm	Concentration of NO after reaction, ppm	NO reduction rate, %
Glass	5—10	371	202	45.55
CaO	11.8	373	195	47.72
MgO	8.2	373	233	37.53
Al_2O_3	12.6	376	194	48.40

It can be seen that when Al_2O_3 is used to be the dielectric, NO reduction rate is the highest.

The intensity E_g of electric field in DBD can be expressed as $E_g = \frac{V_m \varepsilon_g}{2l_d\varepsilon_g + l_g\varepsilon_d}$. In this expression, l_d represents the thickness of the dielectric and l_g represents the breadth of the gas gap. ε_d and ε_g represent the dielectric constant of the dielectric and gas gap respectively. V_m represents the voltage between the plate electrode and electrode tip.

From the above expression, it can be seen that the increase of ε_d can cause the increase of the intensity E_g of electric field. Because the ε_d of Al_2O_3 is the biggest among the 4 dielectrics, NO reduction rate will be the biggest when Al_2O_3 is used to be the dielectric. High dielectric constant ε_d will cause high intensity E_g of electric field.

3 Conclusions

Using the experimental NO reduction system with single-pair electrode tip discharge structure, the NO reduction characteristics of N_2-NO system were revealed to provide a basic reference to the engineering practice, and the results obtained from the NO reduction with single-pair electrode tip discharge plasma also have the same instructive meaning to the NO reduction with dielectric-barrier discharge plasma.

The experimental results of NO reduction with SETD indicate that the amount of both active N atom and NO removal rate increase with the increase of the distance l_g between two electrode tips and then drop when the distance exceeds a certain value, the NO removal rate increases while the voltage V_m between two electrode tips or the resident time of gas flow increases. The distance l_g is a key geometrical variable factor which can determine the intensity E_g of electric field between two electrode tips and the resident time of gas.

The effects of the dielectric features on NO reduction using dielectric-barrier discharge (DBD) plasma system were also studied.

The experimental results of NO reduction rate with different dielectrics such as Al_2O_3, CaO, MgO and glass indicate that the intensity E_g of electric field with different dielectric is different, because it brings different energy to particles in discharge room and thus it causes different NO removal rate. Because the ε_d of Al_2O_3 is the biggest among the 4 dielectrics, NO reduction rate will be the biggest when Al_2O_3 is used. High dielectric constant ε_d will cause high intensity E_g of electric field.

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