Effect of activated NH_3 on SO_2 removal by pulse corona discharge plasma in flue gas

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Abstract: NH_3-activated electrode is placed in front of the electrode system of pulse corona discharge plasma. There are nozzles on the electrode. Positive DC high-voltage is applied on the nozzle-plate gap. NH_3 is injected into the reactor through nozzles, at the same time, activated and treated. These nozzles were proposed in order to make the additional gas pass through corona discharge regions near the tip of nozzles and increase the mount of radicals. The aim is to improve the De-SO_2 efficiency by pulse discharge plasma in flue gas. The following topics are investigated and discussed in the paper: De-SO_2 effect of single NH_3-activated electrode, De-SO_2 effect of activated NH_3, the relationship between stoichiometric ratio of NH_3 to SO_2 and De-SO_2 effect of activated NH_3, mechanism of activated NH_3 De-SO_2 effect. The experimental result indicates that the De-SO_2 efficiency can be increased 5%—10% by activated NH_3 on the original base of De-SO_2 efficiency.

Key words: activated NH_3; De-SO_2; pulse discharge

Introduction

Now SO_2 is the one of main gas pollutants and the main cause of acid rain problem. SO_2 removal by pulse corona discharge is a new De-SO_2 technique developed during last ten years (Masuda, 1993; Mizuno, 1986; Dinelli, 1990). SO_2 in flue gas is oxidized by activated radical (such as OH, HO_2, NH_2, O) which are generated by corona discharge. With NH_3 being injected into the reactor, the final product is ammonium sulfate, which is well known as fertilizer (Wu, 1998). For the oxidized efficiency, the main influence factor is the yield of radical. Recent studies have appeared with investigation on improving the yield of radical (Chang, 1991).

NH_3 has two effects in De-SO_2 process. First, NH_3 acts as additional gas and reacts with SO_2 ammonium sulfate; second, under the condition of corona discharge, NH_3 provides strong oxidized radical, such as NH, NH_2 and so on. However, the stoichiometric ratio of NH_3 to SO_2 is less than or equal to 1:1, the concentration of NH_3 in flue gas is very low. But the collision between high-energy electron and gas molecule is unselective, so the quantity of radical correspond to NH_3 is very small. Thus, promoting the collision between NH_3 molecule and high energy electron is one of the effective methods to improve De-SO_2 efficiency.

NH_3 is injected into the reactor through nozzles and pass through corona discharge. By this method, the yield of activated radical corresponds to NH_3 and the oxidization of SO_2 is increased. Thus, we get the purpose of improving De-SO_2 efficiency.

1 Experimental configuration

The experimental configuration is shown in Fig. 1. The reactor (1) includes activated electrode system and pulse corona discharge electrode system. Activated electrode system consists of nozzle discharge electrode(2) and plate electrode(4); pulse corona discharge electrode consists of line(star-shaped) discharge electrode(3) and plate electrode(4). (5) is a positive high DC power; (6) is a rotate spark-gap pulse power. The schematic diagram of nozzle electrode (2) is shown as Fig. 2. The main body of the electrode is a pipe, 300 mm in length, outside diameter is 10 mm, inside diameter is 8 mm. Nozzle is 30 mm in length, outside diameter 2 mm, inside diameter 1 mm. The distance(D) between nozzle is 50 mm. The plate electrode (4) (500 mm × 900 mm) is

grounded. Air and SO_2 mixture gases act as the simulation gas. The concentration of SO_2 can be adjusted. The simulation gas is heated to 60—$80\,^{\circ}\!C$ and flows into the reactor. The flow rate and the volume ratio of water in flue gas are $10\ m^3h^{-1}$ and 0.8%.

Voltage signal is detected by HP-60 high-voltage probe (Iwasu, Electric Inc. , Japan), current signal by TM502A current probe(Tektronix Inc. , USA). The concentration of SO_2 is measured by IRA-107 SO_2 analyzer(Shimadzu Inc. , Japan).
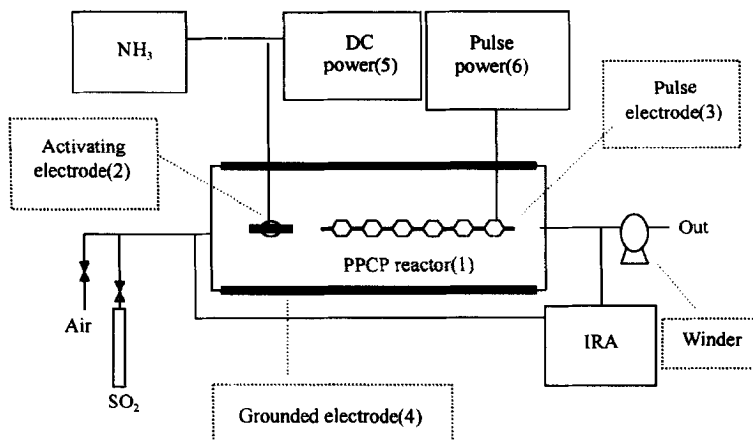

Fig. 1   Experimental configuration

2   Discharge V-I characteristics and pulse waveform of corona discharge

Discharge V-I characteristic in activated electrode system is shown in Fig. 3. It shows that corona current increases with voltage. Thus, the energy in activated field can be calculated. The energy consumption of unit volume flue gas and NH_3 also can be calculated. Typical pulse waveform of corona discharge applied on the reactor is shown in Fig. 4.

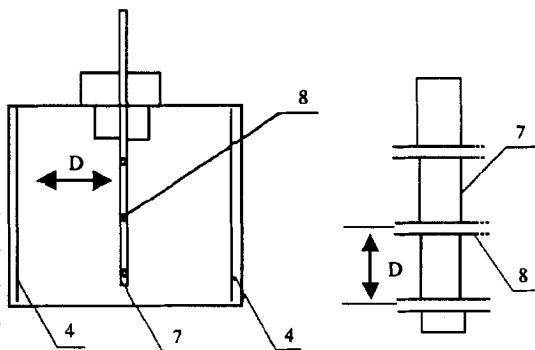Fig. 2   Structure of nozzle electrode

3   Experimental results and discussion

3.1   The effect of activated NH_3 for De-SO_2 efficiency

The stoichiometric ratio of NH_3 to SO_2 is $1:1$, and the initial density of SO_2 is 1760 ppm, no pulsed voltage is applied. Under the condition of activated NH_3, the relation curve of De-SO_2 efficiency and activated voltage is shown in Fig. 5. According to the curve, De-SO_2 efficiency increases with voltage. When the voltage is 28 kV, the De-SO_2 efficiency can be increased by 9% above the original basis of thermal reaction, which is shown as the dotted line($\eta = 66\%$) in Fig. 5. The total removal rate is up to 75%. The increased removal rate due to the following reasons. The electric field intensity is reinforced because of the increased voltage applied to the NH_3-activated electrode. The collision probability between the electrons with high energy and NH_3 molecule is improved. This brings about the increments of energy and the yield of activated NH_3 radicals(NH, NH_2).
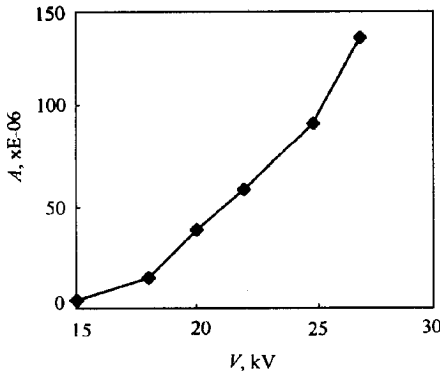
Fig.3   V-I characteristics of corona discharge on
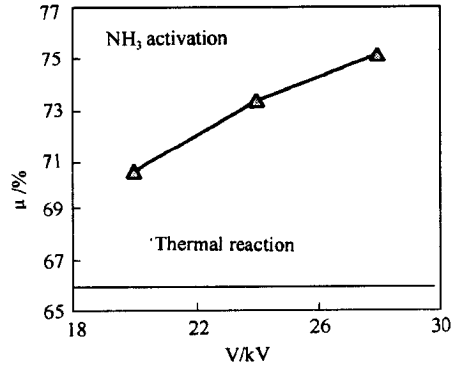activated electrode

Fig.4   Typical pulse waveform of corona
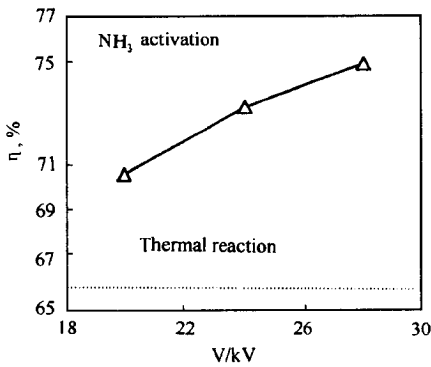discharge

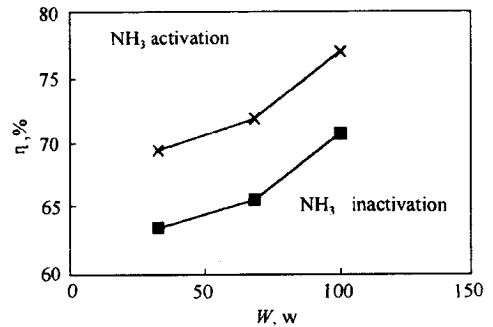Fig.5   Relationship between voltage of NH_3
activation and De-SO_2

Fig.6   Activated effect curve of De-SO_2 pulse
discharge

3.2   The effect of activated NH_3 for SO_2 removal rate in the condition of pulse corona

The stoichiometric ratio of NH_3 to SO_2 is 1:1, the initial density of SO_2 is 1520 ppm, DC high voltage (28 kV) is applied to activate NH_3. Pulse voltage is added on the line electrode. Under the conditions which NH_3 is activated and not, Fig. 6 shows the relationship between De-SO_2 efficiency of pulse corona discharge and pulse energy. According to Fig. 6, De-SO_2 efficiency increases with the pulse energy and under the same energy, De-SO_2 efficiency with activated NH_3 is higher apparently than that NH_3 is not activated. Under the experimental conditions and when activating voltage is 28 kV, De-SO_2 efficiency increase by 6% because of the activated NH_3. There are two reasons. The first is that the pulse energy rises improves the oxidized rate from S^{4+} to S^{6+} in the pulse corona discharge fields; second, NH_3-activated brings conditions to De-SO_2 by pulsed discharge in the later process(the reason of improving De-SO_2 efficiency with activated NH_3 was discussed in 3.1).

3.3   The influence of activated NH_3 quality on De-SO_2 efficiency

The initial density of SO_2 is 1520 ppm. DC voltage is 28 kV. No pulse voltage is applied to NH_3-activated electrode. The stoichiometric ratio of NH_3 to SO_2 is 0.6, 0.8 and 1.0 respectively. Under the two conditions that NH_3 is activated(NH_3 is injected in NH_4-activated electrode) and not (NH_3 is injected into front end of reactor directly). Fig. 7 shows the relationship between De-SO_2 efficiency and R (stoichiometric ratio). The figure indicates that the De-SO_2 efficiency increases with R. When R is the same, the De-SO_2 efficiency with activated NH_3 is clearly higher than that

with NH_3 is not activated. In the same time, the increment between the two lines is improved with R.

Under the conditions of this experiment, when R is 1.0, the improved value of De-SO_2 efficiency can reach 9.5% and the total De-SO_2 efficiency by NH_3 activation and thermal reaction is up to 67%. This is because R is increased, then, the quality of activated NH_3 is increased too. This means more radicals(NH, NH_2) are generated, thus, the De-SO_2 efficiency is increased.

4   Discussion on activation mechanism
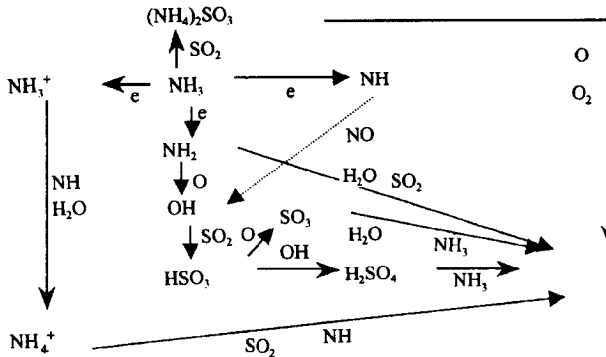
Fig. 8 shows the mechanism of NH_3

Fig.7   Curve of relationship between quality of activated NH_3 and De-SO_2 efficiency

Fig.8   The mechanism of NH_3 activation

activation. When NH_3 is distributed in DC electric field, the collision probability between NH_3 and high-energy electrons, and then, NH_3 is decomposed and ionized. More radicals(such as NH, NH_2, NH_3, OH, etc.) are produced. These radicals react with SO_2 molecules. Thus, SO_2 is transformed to no-polluted substance.

5   Conclusion

(1) De-SO_2 efficiency by NH_3 activation increases with activating voltage. (2) NH_3 activation makes De-SO_2 efficiency increased apparently. (3) The improved value of De-SO_2 efficiency by NH_3-activation effect increases with stoichiometric ratio of NH_3 and SO_2. (4) Under the conditions of this experiment, NH_3 activation effect on De-SO_2 efficiency is up to 5%—10%.

References:

Chang J S, Lawless A, Toshiaki Yamamoto, 1991. IEEE Transaction on Plasma Science[J], 19(6): 1152—1166.

Dinelli G, M Rea, 1990. Journal of Electrostatics[J], (25): 23—31.

Masuda S, S Hosokasa X, T Xu, 1993. International conference on applied electrostatics[C]. Beijing. 1—23.

Mizuno A, J Sidney, Robert H D, 1986. IEEE Trans Ind Appl[J], 22(3): 516—521.

Wu Y, Wang N H, Zhu Y M, 1998. Journal of Electrostatics[J], 44: 11—16.