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An enhancement method for the elemental mercury removal from coal-fired flue gas based on novel discharge activation reactor

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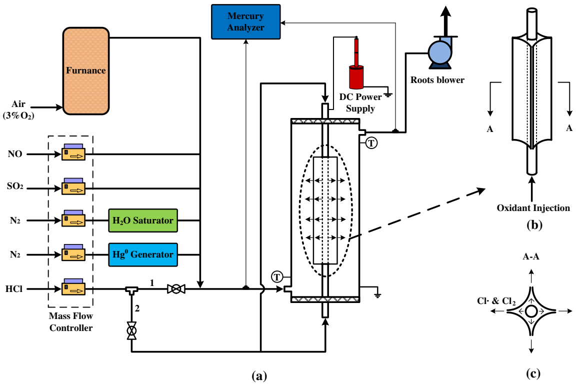
HIGHLIGHTS

- HCl showed a positive effect on the oxidation of Hg^0 in ESP electric field.
- The reaction mechanism between HCl and Hg^0 in ESP electric field was investigated.
- A novel discharge activation reactor was employed to improve Hg^0 removal.

GRAPHICAL ABSTRACT



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ABSTRACT

Gas-phase oxidation of elemental mercury (Hg^0) from flue gas by simulated electrostatic precipitators (ESP) electric field was explored in this paper. In order to enhance the removal efficiency of Hg^0 by ESP electric filed, a novel discharge activation reactor was designed and employed. The influence of HCl concentration, temperature, fly ash, and flue gas components on the Hg^0 removal were also considered, respectively. The Hg^0 removal efficiency increased with the increase of HCl concentration, temperature and discharge voltage. It has also been found that O_2, H_2O and fly ash showed a light promotion on the removal of Hg^0 while NO and SO_2 had a slight inhibition effect on Hg^0 oxidation. Furthermore, the novel discharge activation reactor could improve the generation of reactive chemical species, such as Cl· or Cl_2, which facilitated the mercury removal. At the reaction temperature of 413 K, about 60% Hg^0 could be removed from simulated flue gas under 25.0 kV in presence of 10.0 ppmv HCl. When the novel discharge activation reactor was used, the Hg^0 removal efficiency increased to about 80% at the same experimental conditions. It appeared to be a promising technique to enhance the removal of Hg^0 by ESP.

1. Introduction

The Minamata Convention on Mercury, the world's first legally binding treaty which aiming at reducing the mercury pollution has been adopted in 2013. As a toxic persistent bioaccumulative pollutant, mercury pollution has become a worldwide environmental problem and received unprecedented attention [1,2]. Coal-fired flue gas is considered as one of the main anthropogenic sources, it is significant to control the mercury emission from coal-fired flue gas [3]. Among the three main forms of mercury in flue gas, elemental mercury (Hg^0) is more difficult to be removed than oxidized mercury (Hg^{2+}) and particulate-bound mercury (Hg^P)

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because of its infusibility and highly volatility [4]. Furthermore, Hg^0 accounts for about 20–70% of the total mercury in flue gas [5,6]. Therefore, how to remove Hg^0 effectively is the key to control the mercury emission from coal-fired flue gas [7].

The existing air pollution control devices (APCDs) could be used to reliably provide a high level of mercury control [8]. It is a tendency to remove mercury by using APCDs which are widely applied in coal-fired power plants and very effective to remove Hg^{2+} and Hg^P [9,10]. Accordingly, many researches have been focused on the converting Hg^0 into Hg^{2+} species to make full use of APCDs in mercury removal from flue gas.

Many techniques, such as oxidant injection [11,12], catalytic oxidation [13,14], non-thermal plasma (NTP) [15,16], have been studied on the oxidation removal of Hg^0. However, the instability of injected oxidants limited the practical applications of chemical oxidant injection [16]. Meanwhile, the fly ash and SO_2 in flue gas will inhibit the Hg^0 removal by catalyst [17]. As an environmental friendly air pollutant control technology, the NTP have been extensive studied in decades. In NTP process, electrical discharge will generate many free electrons and reactive chemical species, such as $\cdot OH$, O, O_3 and $\cdot O_2H$. These active chemicals supply the electrical discharge with a unique chemical environment and are able to oxidize elemental mercury effectively [18,19]. Compared with the NTP, the electrostatic precipitator (ESP) electric field could also generate many radicals and gas ions which may react with Hg^0 and form Hg^{2+} [20]. Moreover, the halation produced in the ESP discharge process will be helpful for the Hg^0 oxidation. Furthermore, ESP has been widely used in the air pollution control. It seems to be a good choice to remove mercury by ESP. However, it was reported that the average mercury removal efficiency by the ESP was about 29%, which can hardly meet the increasingly stringent requirements of mercury emission control [21]. To the best of our knowledge, there is few study about enhance mercury oxidation by ESP electrical discharge so far. In order to improve the mercury removal by ESP, the oxidation of Hg^0 and its enhancement method in simulated ESP electric field was preliminary investigated in this paper. The impact of HCl concentration, reaction temperature, discharge voltage and different flue gas components have been fully considered, respectively.

2. Materials and methods

2.1. Experimental setup and procedures

Three simulated ESP reactors were used in this research. One bench-scale static reactor (reactor A) in Fig. S1 was used to study the reaction kinetics between Hg^0 and HCl under electric field. It was made of a stainless steel cylinder with a volume of 1100 mL. The reactor cover was made of polytetrafluoroethylene (PTFE). A serrated stainless steel was fixed in the center of the reactor cover which was 30 mm width and connected with the anode of high voltage power supply. The bottom of reactor was connected with ground as the cathode. The distance between the top of anode serrate and the cathode is about 30 mm. High voltage (from 1.5 kV to 10.5 kV) is applied between two electrodes. The inner wall of the reactor was coated with halocarbon wax to minimize the effect of wall in the reaction of Hg^0 with HCl. The concentration of Hg^0 in the reactor was continuously on-line detected by a cold vapor atomic absorption spectrophotometer (CVAAS).

Meanwhile, a bench-scale dynamic reactor (reactor B) in Fig. S2 was used to study the Hg^0 removal in fluidized flue gas. This dynamic reactor is a glass cylinder with its inner diameter of 27 mm and length of 150 mm. Anode is a serrated stainless steel which is fixed vertically in the top of reactor. Cathode is placed horizontally in the reactor which is rectangle stainless steel. The length, width and thickness of both electrodes are 35 mm, 10 mm and 1 mm, respectively. The distance between the top of anode serrate and the cathode is about 16 mm. The volume of the discharge area in reactor is about 20 mL. The reactor is placed in an oven to maintain the simulated flue gas temperature from 303 to 413 K. High voltage (from 1.5 kV to 10.5 kV) is applied between two electrodes. The concentration of Hg^0 was about $20.0 \pm 0.5\ \mu g/m^3$. The simulated flue gas flow was 40.0 ± 1.0 L/h.

Finally, a pilot-scale ESP reactor (reactor C) was used to study the performance of novel discharge activation reactor in Hg^0 removal from fluidized flue gas as shown in Fig. 1. This pilot-scale dynamic reactor is a stainless steel tube with inner diameter of 150 mm and length of 2000 mm. The anode consists of a hollow stainless prism and a porous stainless pipe (Fig. 1b). The pipe passes through the inner cavity of prism along the axis direction and is welded on the prism. The intersecting surface of the prism is starlike and its four edges are cracks with the width of 0.5 mm (Fig. 1c). When the oxidant is injected into the reactor through the anode, it will enter the reactor through the hole of inside pipe and the cracks of prism. The length of the anode is 1500 mm. The cathode is the wall of stainless steel tube which is connected with the ground. The distance between the edge of prism anode and the cathode is about 50 mm. The flue gas temperature of reactor is maintained at 413 K through an electric heating furnace. High voltage (from 5.0 kV to 30.0 kV) was applied between two electrodes. A gas mixture containing N_2, O_2, H_2O, HCl, SO_2, NO and fly ash was used to simulate flue gas. The concentration of O_2, H_2O, HCl, SO_2, NO and fly ash is 3.5%, 1.0%, 10.0–80.0 ppmv, 500.0 ppmv, 500.0 ppmv and 25.0 g/m^3, respectively. The concentration of Hg^0 source was kept at $35.0 \pm 1.0\ \mu g/m^3$. The simulated flue gas flow was $10.0 \pm 1.0\ m^3/h$.

2.2. Chemicals

The chemicals used in this research are mercury (99.99%) from Sigma–Aldrich Co. N_2 (99.99%), SO_2 (10.0%), NO (10.0%), O_2 (99.9%) and HCl (10.0%) are from Dalian Date Standard Gas Co. Specific detecting tubes for SO_2, NO and HCl measurement from Gastec Co. In the tests, the trace gases of SO_2, NO and HCl were 500 ppmv, 500 ppmv, and 10 ppmv, respectively. And the carrier gas was N_2.

2.3. Analytical methods

Hg^0 was continuously monitored by a mercury analyzer (SG-921, Jiangfen, China) which based on cold vapor atomic absorption photometry (CVAAS). The signal was collected with a data transition and acquisition device (N2000, Zhida, China) and recorded by computer. The CVAAS signal was calibrated by a mercury analyzer (RA-915, Lumex, Russia). In order to accurately assess and report the measurement uncertainties for this work, each experiment of Hg^0 removal efficiency was carried out at least three times. The Hg^0 removal efficiency (η) was defined as:

$$\eta = \frac{[Hg^0]_0 - [Hg^0]_t}{[Hg^0]_0} \times 100\% \tag{1}$$

where $[Hg^0]_0$ and $[Hg^0]_t$ are the concentrations of Hg^0 at before and after treatment, respectively.

In order to deduce the reaction mechanism between HCl and Hg^0 in ESP electric field, the decomposition process of HCl in ESP electric field was investigated. 10% HCl was injected into the static reactor, and the discharge voltage was kept at 9.0 kV. The reaction products was determined and recorded by a UV/vis spectrometer (BRC641E, BWTEK, USA).

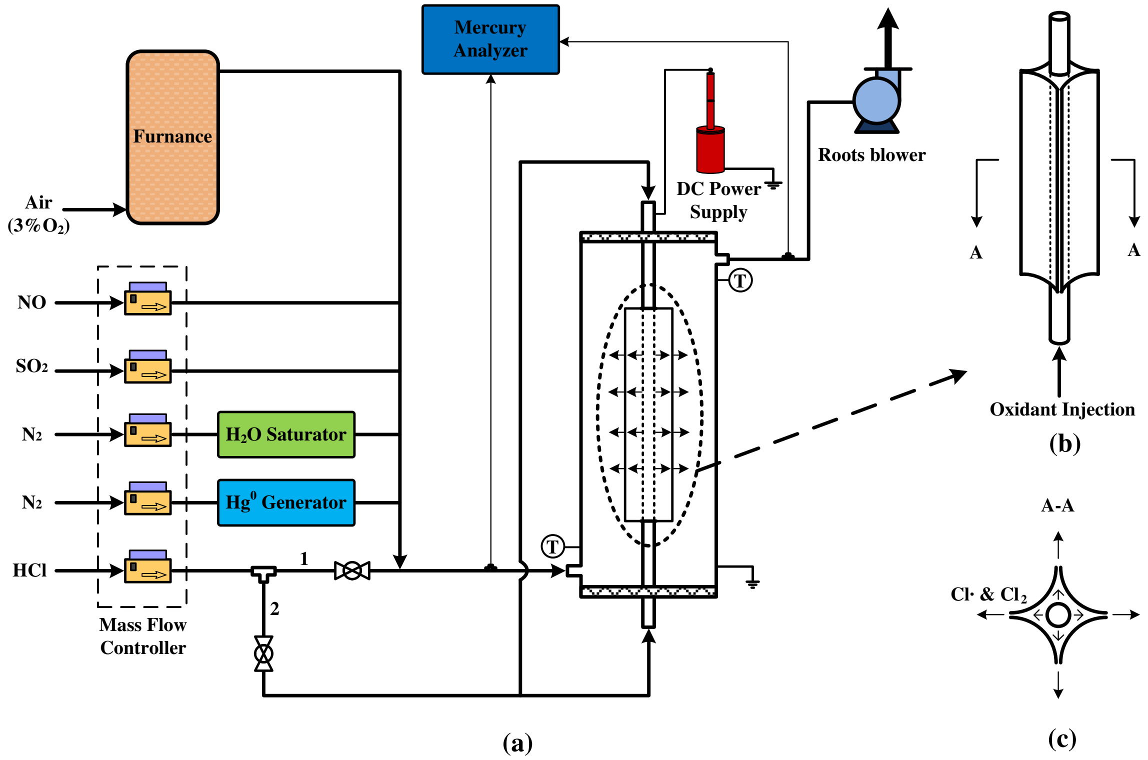

Fig. 1. Schematic diagram of the pilot-scale apparatus for Hg^0 oxidation in simulated ESP electric field.

3. Results and discussion

3.1. Hg^0 oxidation in the presence of HCl in ESP electric field

The chlorine species, such as HCl and Cl_2, are considered to be the major Hg^0 oxidation reagents in coal-fired flue gas. Many researches have been conducted to use chlorine species remove Hg^0 from flue gas. Chemical kinetic models suggest that the chlorine content of the flue gas is the main contributor to mercury oxidation [22]. Increasing the concentration of HCl in flue gas will promote $HgCl_2$ formation [23]. But the reactivity of HCl toward Hg^0 is too low to meet the increasingly stringent requirements for Hg^0 removal from flue gas [24]. In this study, ESP electric field is introduced to enhance the Hg^0 removal by chlorine species.

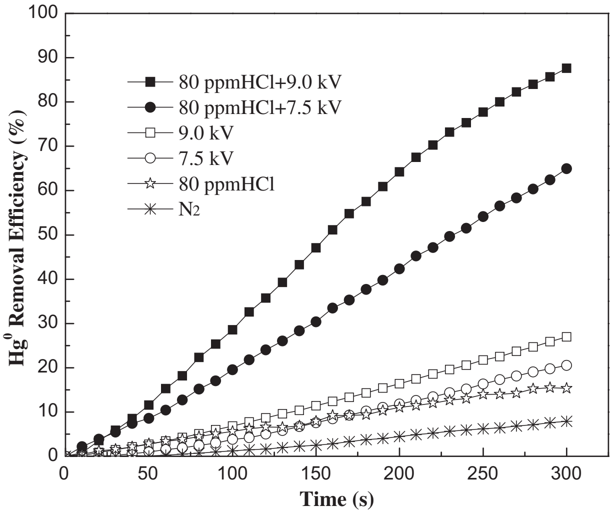

Fig. 2. Effect of HCl and high voltage discharge on Hg^0 removal.

Fig. 2 showed that the removal efficiency of Hg^0 in the reactor A was no more than 20% at 300 s when the HCl concentration was 80.0 ppmv or the voltage was 9.0 kV. It indicated that the Hg^0 couldn't be removed effectively only by HCl or electric field with high voltage discharge. While, the removal efficiency of Hg^0 increased to 87.6% after 80.0 ppmv HCl was injected into the reactor and voltage was increased to 9.0 kV simultaneously. Obviously, the Hg^0 can be removed effectively by the combination of HCl and high voltage discharge. In the reactor A, the main components included N_2, HCl and Hg vapor. Other impurity components were extremely low, which affecting the reaction system could be ignored. Therefore, the reaction rate constant was calculated according to the concentrations of HCl and mercury.

As can be seen in Fig. 2, HCl under high voltage was high effective on Hg^0 oxidation. When the HCl was kept around 80 ppmv, the oxidation efficiencies at a residence time of 150 s were about 30% and 47% for HCl at 7.5 kV and HCl at 9.0 kV, respectively. To further analyze the data in Fig. 2, it appears a pseudo-first-order rate kinetics between Hg^0 and the oxidants. The oxidation rate of Hg^0 also appears to be directly proportional to the oxidant concentration. Therefore, it can be concluded that the gas-phase reaction of Hg^0/HCl under high voltage is second-order with respect to Hg^0 and HCl, as expressed by Eq. (2):

$$-d[Hg^0]/dt = k_2[Hg^0][HCl] \tag{2}$$

where $[Hg^0]$ is the concentration of Hg^0 in the gas phase in molecules/mL, and k_2 is the second-order rate constant. The rate constant from our data is:

$1.97(\pm 0.05) \times 10^{-18}$ mL/molecules/s for HCl at 9.0 kV and

$1.11(\pm 0.05) \times 10^{-18}$ mL/molecules/s for HCl at 7.5 kV at 373 ± 2 K and 760 Torr. The rate constant of Hg^0 oxidation by HCl under high voltage is more than one order of magnitude larger than that by Cl_2 [33]. It can be seen that the k_2 increased with the increase of the discharge voltage in the reactor.

3.2. Influence of reaction temperature and discharge voltage on Hg^0 removal

The influence of reaction temperature and discharge voltage on Hg^0 removal by HCl and ESP electric field was also investigated through the reactor B. From Fig. 3 it can be seen that the removal efficiency of Hg^0 increased from 32.5% to 87.8% with the reaction temperature increased from 303 to 413 K. The initial voltage needed is lower when the temperature is higher. That may because the higher temperature could provide more energy which facilitates the generation of Cl atom or Cl_2. Generally, the temperature of flue gas in ESP is between 393 and 453 K. Thus, the real flue gas temperature is favored for the Hg^0 removal by HCl and ESP electric field.

3.3. Discussion on the reaction mechanism in ESP electric field

In order to investigate the enhancement effect of HCl and electric field on mercury removal, the reaction product of HCl in high voltage discharge process was determined and recorded by UV/vis spectrometer. As shown in Fig. 4, there was not obvious absorption peak at the beginning of reaction. After 25 min, an increasing absorption peak occurred at about 330 nm. The maximum UV adsorption peak of chlorine is at approximately 330 nm [11]. In order to facilitate testing Cl_2, the HCl concentration was increased to 10%. Although Cl_2 was not detected in 5 min, actually there has been a part of HCl into chlorine. Therefore, it was believed that Cl_2 molecules and Cl atoms were generated in high voltage discharge process. In high voltage discharge process, electrons produced by electrical energy collisions with N_2 will generate excited N_2 molecules (N_2^*) [25]. The electrons and active species will promote the formation of Cl or Cl_2. Then, these chlorine species will react with Hg^0. All of these reactions are presented as following:

$$HCl + N_2^* \rightarrow H + Cl + N_2 \qquad (a)$$

$$HCl + e \rightarrow H + Cl + e \qquad (b)$$

$$HCl + H \rightarrow H_2 + Cl \qquad (c)$$

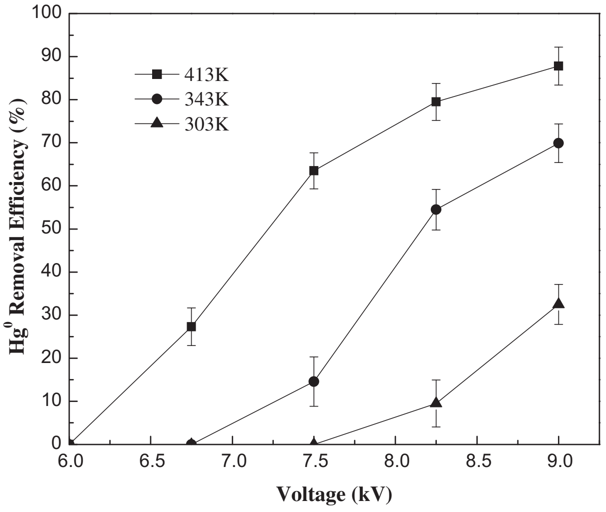

Fig. 3. Influence of flue gas temperature on Hg^0 removal.

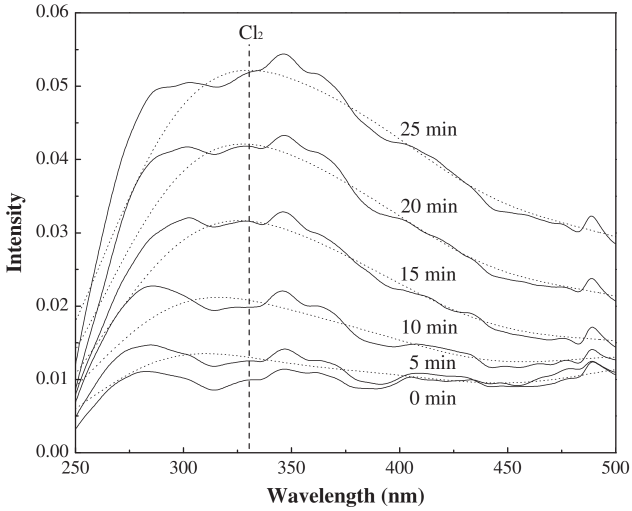

Fig. 4. Spectrum evolution for the decomposition of HCl in ESP electric field.

$$Cl + Cl \rightarrow Cl_2 \qquad (d)$$

$$Hg^0 + Cl \rightarrow HgCl \qquad (e)$$

$$HgCl + Cl \rightarrow HgCl_2 \qquad (f)$$

$$Hg^0 + Cl_2 \rightarrow HgCl_2 \qquad (g)$$

The reaction between Hg^0 and HCl is unlikely to proceed via the route of gas-phase reaction because this reaction is hindered by high activation energy [26]. But the reaction rate constant of Hg^0 and Cl is very large [27]. Thus, the decomposition of HCl in electric field will promote the Hg^0 removal. It is kinetically and thermodynamically more favorable to produce HCl by the recombination of H and Cl atoms, rather than forming H_2 and Cl_2 [25]. According to the data from Fig. 4, it can be calculated that the concentration of Cl_2 is only about 760.0 ppmv after 25 min reaction. Obviously, the productivity of Cl_2 was too low (less than 0.8%) to remove Hg^0 effectively. If the yield of chlorine activity species in ESP electric field could be improved, the Hg^0 removal efficiency will also be increased. Therefore, there is great potential to enhance the Hg^0 removal by developing the synergistic effect of ESP electric field and HCl.

3.4. Hg^0 removal in novel discharge activation reactor

A pilot-scale ESP reactor was used to investigate the Hg^0 removal in the electric field which closed to the actual situation in industrial application. The removal efficiency of Hg^0 as a function of voltage was obtained at different HCl concentration in N_2. As shown in Fig. 5, the Hg^0 removal efficiency increased from 61.5% to 80.3% at 25.0 kV when HCl concentration increased from 10.0 ppmv to 80.0 ppmv. The Hg^0 removal efficiency was improved by near 20% at the high voltage. While, the removal efficiency of Hg^0 decreased a little when the discharge voltage increased to 30.0 kV and the electrical breakdown happened subsequently. When the HCl concentration increased from 80 ppmv to 120 ppmv, the removal efficiency of Hg^0 improved not much. This indicated that there was a limit to Hg^0 removal by HCl concentration. The HCl concentration of coal-fired flue gas is about 10.0–100.0 ppmv [25]. It should be enough for the mercury removal by ESP. Nevertheless, the mercury removal performance of ESP wasn't satisfied. It is probably because only part of the HCl molecules, which near the discharge electrode, could be activated by ESP electric field.

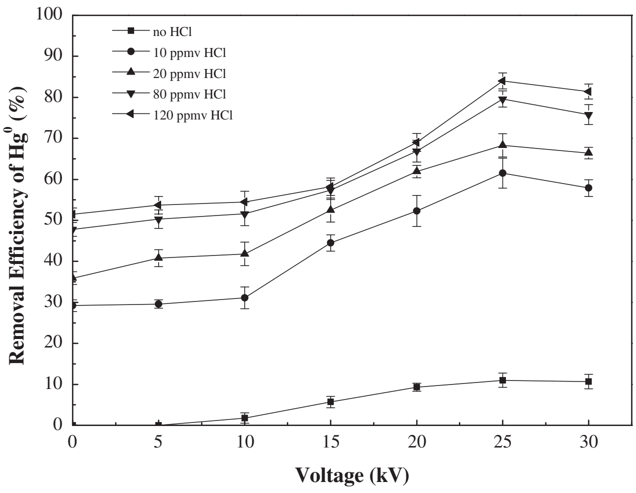

Fig. 5. Influence of HCl concentration on Hg^0 removal.

So the chlorine activity species generated are not enough for the mercury removal accordingly. Huge amount of HCl gas should be injected into the flue gas to enhance the mercury removal by ESP. For the sake of reducing the usage of HCl gas and increasing the yield of chlorine activity species in ESP field, we have designed a novel HCl discharge activation reactor which showed as in Fig. 1b. In this research, HCl gas was injection into the inside pipe of anode (inside channel). Then it entered the reactor through the cracks of prism. Compared with the traditional oxidant injection style (outside channel), there are two major benefits to inject the HCl through the inside channel. Firstly, all of the injected HCl gas will pass the discharge area of ESP because it had to enter the reactor through the cracks of anode prism. Secondly, high concentration HCl gas (up to 100%) could be injected through the inside channel. Higher concentration HCl gas has more chance to be activated by electric field. Therefore, the productivity of chlorine activity species and the removal efficiency of Hg^0 will be improved greatly. In order to evaluate the enhancement performance of the discharge activation reactor, HCl gas was injected into the flue gas through different channels. As shown in Fig. 6, the Hg^0 removal efficiencies were about 61.5% and 79.6% when the HCl gas was injected through the outside channel and inside channel, respectively. The amplification of Hg^0 removal efficiency was about 18.1% after the discharge activation reactor was used. Since the operational conditions in real ESP devices, such as the equipment structure, discharge voltage and residence time, are better than that in this research, the Hg^0 removal efficiency was expected to be higher in the actual working conditions.

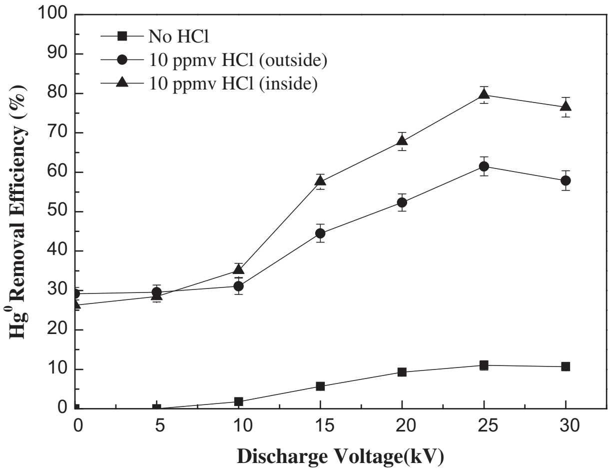

Fig. 6. Comparison test result of the before and after improvement.

3.5. Influence of flue gas components on Hg^0 removal

The flue gas composition is very complex. The main components of flue gas are N_2, CO_2, O_2, H_2O, NO, SO_2 and fly ash. These components may affect the removal of Hg^0 in ESP electric field. Therefore, the influence of these major components was investigated in this part. The removal efficiency of Hg^0 with different gas components in ESP electric filed was shown in Fig. 7.

From Fig. 7, CO_2 appeared to be very inert in this environment, and no effect was observed in such processes. Hg^0 removal efficiency in 10% CO_2/N_2 at different discharge voltage was shown in Fig. S4. The result indicated that CO_2 concentration showed insignificant effect on the Hg^0 removal. O_2, H_2O and fly ash showed a light promotion on the removal of Hg^0 by ESP electric field in presence of HCl. In high voltage discharge process, most of the electrical energy went into the production of energetic electrons [28]. These electrons collide with O_2 or H_2O and lead to the formation of O, O_3 and $\cdot OH$. Eventually, these chemically active species will react with Hg^0 [29]. Fly ash promoted the mercury adsorption. The adsorption of mercury by fly ash included physical and chemical adsorption. Fly ash had the promoting effect on the Hg^0 removal [1,22,34]. However, the removal efficiency of Hg^0 was not high. Therefore, the focus was to investigate how to improve the mercury removal efficiency by ESP, namely develop a new activation device using discharge and HCl synergy to generate highly active chlorine atoms to strengthening the gas phase oxidation efficiency of Hg^0 in this paper. As can be seen in Fig. 7, NO and SO_2 had a slight inhibition effect on Hg^0 oxidation. It's believed that NO and SO_2 can rapidly react with the active species Cl atoms and Cl_2 produced by ESP electric field, thus the Hg^0 oxidation reaction was inhibited. The correlative reaction mechanism showed as following [30–32]:

$$Cl_2 + NO \rightarrow NOCl + Cl \tag{h}$$

$$Cl + NO \rightarrow NOCl \tag{i}$$

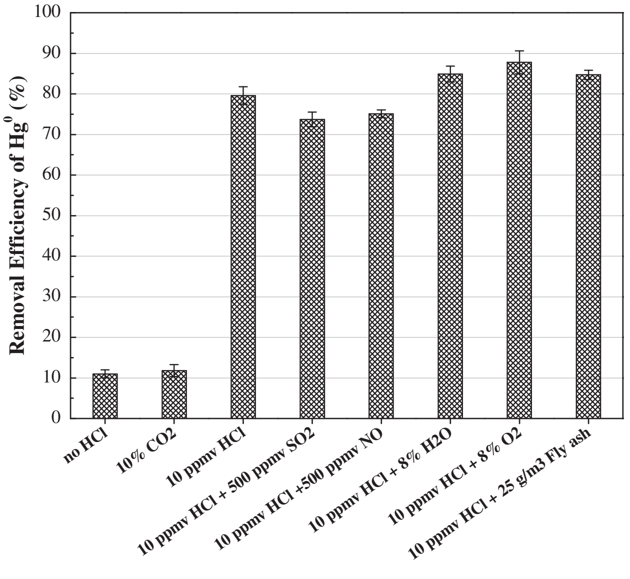

Fig. 7. Influence of flue gas compositions on Hg^0 removal.

$$Cl_2 + SO_2 \rightarrow SO_2Cl_2 \tag{j}$$

$$Cl + SO_2 \rightarrow SO_2Cl \tag{k}$$

The formed SO_2Cl, SO_2Cl_2, NOCl, and $NOCl_2$ are capable of decomposing back to Cl and Cl_2 or have nearly an equal oxidizing property as Cl and Cl_2, respectively [32]. Thus, SO_2 and NO cannot substantially inhibit Hg^0 oxidation.

4. Conclusions

In conclusion, HCl has a synergistic effect on the Hg^0 removal by simulated ESP reactor. The mercury removal efficiency increased with the increase of HCl concentration, reaction temperature and discharge voltage. The chlorine activity species produced in high voltage discharge process maybe the main reason that improving the gas-phase oxidation of Hg^0 in ESP electric field. But the productivity of chlorine activity species in ESP electric field wasn't satisfied. In order to increase the yield of chlorine activity species in ESP field, a novel HCl discharge activation reactor was designed and tested. The results shown that the Hg^0 removal efficiency increased near 20% after the discharge activation reactor was applied. In addition, the influences of flue gas components on mercury removal performance of this discharge activation reactor were also investigated. O_2, H_2O and fly ash showed a light promotion on the removal of Hg^0. NO and SO_2 had a slight inhibition effect on Hg^0 oxidation by ESP electric field in presence of HCl. Since ESP devices have been used widely in coal-fired power plants, it could be a promising control technique for Hg^0 removal from flue gas.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.fuel.2015.12.066.

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