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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<sup>0</sup> in ESP electric field.
- The reaction mechanism between HCl and Hg<sup>0</sup> in ESP electric field was investigated.
- A novel discharge activation reactor was employed to improve Hg<sup>0</sup> removal.

# G R A P H I C A L A B S T R A C T



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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<sub>2</sub> 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<sub>2</sub>, 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

\* Corresponding authors. Tel./fax: +86 21 54745591. E-mail addresses: quzan@sjtu.edu.cn (Z. Qu), nqyan@sjtu.edu.cn (N. Yan). 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<sup>0</sup>) is more difficult to be removed than oxidized mercury (Hg<sup>2+</sup>) and particulate-bound mercury (Hg<sup>P</sup>) 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<sup>0</sup>. However, the instability of injected oxidants limited the practical applications of chemical oxidant injection [16]. Meanwhile, the fly ash and SO<sub>2</sub> in flue gas will inhibit the Hg<sup>0</sup> 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<sub>3</sub> and  $\cdot$ O<sub>2</sub>H. 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<sup>0</sup> and form Hg<sup>2+</sup> [20]. Moreover, the halation produced in the ESP discharge process will be helpful for the Hg<sup>0</sup> 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<sup>0</sup> 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<sup>0</sup> was about 20.0 ± 0.5  $\mu$ g/m<sup>3</sup>. 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<sup>0</sup> removal from fluidized flue gas as shown in Fig. 1. This pilotscale 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<sub>2</sub>, O<sub>2</sub>, H<sub>2</sub>O, HCl, SO<sub>2</sub>, NO and fly ash was used to simulate flue gas. The concentration of O<sub>2</sub>, H<sub>2</sub>O, HCl, SO<sub>2</sub>, 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<sup>3</sup>, respectively. The concentration of Hg<sup>0</sup> source was kept at  $35.0 \pm 1.0 \,\mu\text{g/m}^3$ . The simulated flue gas flow was  $10.0 \pm 1.0 \text{ m}^3/\text{h}$ .

#### 2.2. Chemicals

The chemicals used in this research are mercury (99.99%) from Sigma–Aldrich Co. N<sub>2</sub> (99.99%), SO<sub>2</sub> (10.0%), NO (10.0%), O<sub>2</sub> (99.9%) and HCl (10.0%) are from Dalian Date Standard Gas Co. Specific detecting tubes for SO<sub>2</sub>, NO and HCl measurement from Gastec Co. In the tests, the trace gases of SO<sub>2</sub>, NO and HCl were 500 ppmv, 500 ppmv, and 10 ppmv, respectively. And the carrier gas was N<sub>2</sub>.

## 2.3. Analytical methods

Hg<sup>0</sup> 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<sup>0</sup> removal efficiency was carried out at least three times. The Hg<sup>0</sup> removal efficiency ( $\eta$ ) was defined as:

$$\eta = \frac{[\text{Hg}^0]_0 - [\text{Hg}^0]_t}{[\text{Hg}^0]_0} \times 100\%$$
<sup>(1)</sup>

where [Hg<sup>0</sup>]<sub>0</sub> and [Hg<sup>0</sup>]<sub>t</sub> are the concentrations of Hg<sup>0</sup> at before and after treatment, respectively.

In order to deduce the reaction mechanism between HCl and Hg<sup>0</sup> 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).

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