

# Mechanism Research of Hg<sup>0</sup> Oxidation by Pulse Corona Induced Plasma Chemical Process

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**Abstract:** The oxidation of Hg<sup>0</sup> by Pulse Corona Induced Plasma Chemical Process (PPCP) was investigated through changing discharge voltage, pulse frequency and gas compositions. Experimental results indicate that active radicals including O, O<sub>3</sub> and OH can contribute to the oxidation of elemental mercury. 10 kV is the onset voltage, and the higher voltage the better removal efficiency. While with the increase of pulse frequency, the Hg<sup>0</sup> concentration falls rapidly at first but then rises rapidly. The best oxidation condition is at 12 kV and 600-800 PPS. Adding O<sub>2</sub> can significantly promote oxidation. With NO and SO<sub>2</sub> existed, there is an inhibition of mercury oxidation, and NO has a greater influence. Addition of HCl can promote oxidation slightly but affect the initial concentration of mercury significantly. Little moisture content can promote oxidation, while too much H<sub>2</sub>O can not only resist the oxidation, but also affect the initial concentration of mercury. The mercury oxidation rate can increase to 97.95% at 12 kV/800 PPS with the system of 10% O<sub>2</sub>/3% H<sub>2</sub>O/50 ppm HCl. However, mercury oxidation efficiency can reduce down to 20% with 100 ppm NO added.

**Key words:** Non-thermal plasma, mercury, oxidation, flue gas cleaning.

## 1. Introduction

Anthropogenic emission of mercury is of great environmental concern because Hg is highly toxic, persistent, bio-accumulative and can cause neurological damage. Out of various sources of mercury pollutants, coal combustion has been considered as one of the largest producers of anthropogenic mercury [1]. As one of the largest coal producers and consumers in the world, China accounts for 42.8% of the coal combustion in the world and releases large amounts of Hg. Chinese government has paid much attention to Hg emission and mandated that the emission of mercury and its compounds should be less than 0.03 mg/Nm<sup>3</sup> after 2015 [2]. Mercury in coal combustion flue gases exists in three forms: elemental (Hg<sup>0</sup>), oxidized (Hg<sup>2+</sup>) and particulate bound (Hg<sup>p</sup>). It is estimated that 70% of Hg was released as gaseous mercury and 20% as Hg<sup>p</sup> in the coal combustion flue

gas [3]. Hg<sup>2+</sup> and Hg<sup>p</sup> can be effectively removed by the conventional air pollution control systems such as Electrostatic Precipitator (ESP), fabric filters and Wet Flue-Gas Desulphurization (WFGD) [4]. However, Hg<sup>0</sup> is difficult to remove because of its high vapor pressure, high volatile and low water solubility [5]. Recently, several techniques have been used in attempts to reduce Hg<sup>0</sup> emission, such as Powder Activated Carbon (PAC) injection, catalytic oxidation and oxidant injection, which usually suffers from the problems of high cost and high carbon-to-mercury ratio of the PAC injection, the deactivation of the catalysts and the instability of the injected oxidants [2]. Therefore, it is imperative to develop cost-effective and highly efficient approaches to oxidize Hg<sup>0</sup> in mercury related industries.

Pulse corona induced Plasma Chemical Process (PPCP) is regarded as a promising technology to control pollutants. In PPCP, reactive chemical species such as OH, O, O<sub>2</sub>H and O<sub>3</sub> are generated by electrical discharge and they lead to the oxidation of Hg<sup>0</sup> to

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Hg<sup>2+</sup> [6]. Short discharge time (nanoseconds), high peak voltage and low power consumption make it possible to realize the goal of low cost and high efficient for pollutants removal. At the same time, the plasma reactor has a simple structure, starts quickly and covers a smaller area, which does not produce secondary pollution and can realize the coordinated control of multiple pollutants [7, 8]. A lot of researches have done at home and abroad about pollutants removal by Non-Thermal Plasma (NTP), especially at desulfurization and denitration and dealing with Volatile Organic Compounds (VOCs) [9-12], which have made certain progress. However, the treatment of heavy metal pollutants is rarely reported. Concentration of mercury in flue gas is relatively low, so there is hope to achieve higher removal efficiency. This article focuses on pulse corona plasma oxidation mechanism and the influence factors of elemental mercury oxidation, eventually realizes the simultaneous removal of SO<sub>2</sub>/NO/Hg<sup>0</sup> by PPCP.

## 2. Material and Methods

A schematic diagram of the experimental system is shown in Fig. 1, which mainly consists of a plasma reactor with high-voltage pulse power supply, mercury generator, water vapor generator, air

distribution system and mercury analyzer. The reactor is wire-in-tube pulsed corona reactor. Flows of different gases, such as N<sub>2</sub>, O<sub>2</sub>, NO, SO<sub>2</sub>, HCl etc. are precisely controlled by individual mass flow controllers. The pure N<sub>2</sub> is sent to pass through the mercury generator to carry out pre-calibrated elementary mercury to the gas stream. Hg<sup>0</sup> concentration was set by controlling the flow rate of N<sub>2</sub>. The mercury generator consists of an Hg<sup>0</sup> permeation tube placed in the center of a “U” type glass tube, with water bath heating system. The mercury analyzer is VM3000 mercury concentration analyzer, produced by MI company in Germany. The principle of the analyzer is based on atomic absorption spectrum method, and the accuracy can reach 0.1 μg/Nm<sup>3</sup>. A large amount of O<sub>3</sub> produced in the plasma reactor, and a single end tail tube furnace heated to 350 °C should be used here to decompose the remaining ozone, so as to eliminate its interference on mercury measurement, since ozone has a similar absorption as Hg<sup>0</sup> at 253.7 nm. The Hg<sup>0</sup> oxidation efficiency is defined in Eq. (1),

$$\eta (\%) = \frac{C_{on} - C_{off}}{C_{off}} \times 100\% \quad (1)$$

Where, C<sub>on</sub> and C<sub>off</sub> are the concentration of Hg<sup>0</sup> (μg/Nm<sup>3</sup>) measured at the outlet of plasma reactor with or without high voltage discharge.

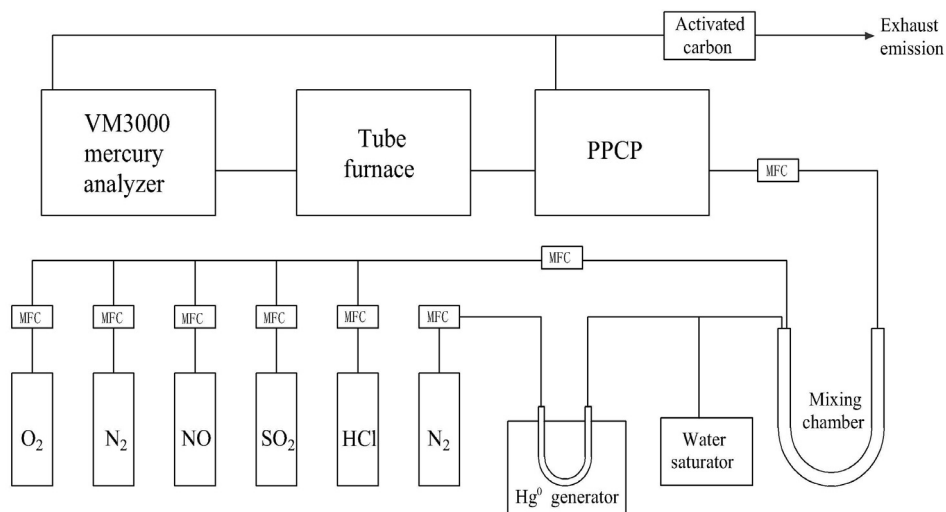


Fig. 1 Schematic diagram of experimental setup for the oxidation of Hg<sup>0</sup> (MFC means “Mass Flow Meter” and PPCP means “Pulse corona induced Plasma Chemical Process” in the diagram above).

### 3. Results and Discussion

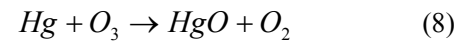
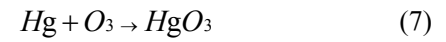
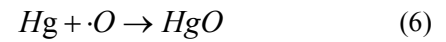
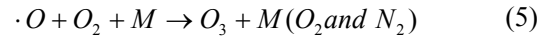
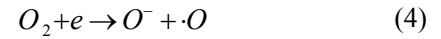
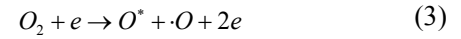
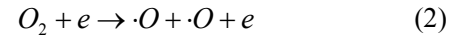
#### 3.1 Mercury Oxidation by Non-thermal Plasma

In order to validate the performance in mercury oxidation, only mercury, N<sub>2</sub> and O<sub>2</sub> appeared in the gas stream during the PPCP with different discharge voltages and different pulse frequency.

The original gas composition was kept as approximately 325 μg/Nm<sup>3</sup> Hg<sup>0</sup> and 33 μg/Nm<sup>3</sup> Hg<sup>0</sup>, with N<sub>2</sub> as carrier, 10% O<sub>2</sub> and N<sub>2</sub> as balance. Fig. 2 shows the variation of Hg<sup>0</sup> concentration with different discharge voltage during the pulse corona induced plasma discharge. When the discharge voltage is lower than 10 kV, Hg<sup>0</sup> concentration is dramatically stable. While, when the discharge voltage is higher than 10 kV, there is a sharp decrease of Hg<sup>0</sup> concentration. It is meaning that the onset voltage of pulse corona induced plasma is about 10 kV, which was similar to the results reported by Zhu, T. L. [13, 14] from Beihang University, China. As shown in Fig. 2, the discharge voltage was 12 kV, Hg<sup>0</sup> concentration decreased to 47 μg/Nm<sup>3</sup> and 3.8 μg/Nm<sup>3</sup>, respectively. The mercury oxidation rate was about 85.5% and 88.5% correspondingly.

The oxidation of Hg<sup>0</sup> here can be ascribed to reactive radicals such as O<sub>3</sub>, O, etc., which were produced in the NTP process through electrons. The oxidative reactive reaction Eqs. (2)-(8) proposed in

literatures [15, 16] can be used to explain the oxidation of Hg<sup>0</sup> in plasma:



In Fig. 3, when the plasma reactor is opened, there is a sharp decrease of Hg<sup>0</sup> concentration. With the increase of pulse frequency, the Hg<sup>0</sup> concentration falls rapidly at first but then raises rapidly. When the frequency is 600-800 PPS, the elemental mercury removal efficiency is the best, which can achieve 95%. While considering the energy consumption problems, 600 PPS is the best choice. He, L. J. [17] also found that with the increase of frequency, toluene removal efficiency increased at first and then decreased with the increase of pulse frequency, which resulted from the situation that when gases discharge, the plasma reactor is a capacitive load. Besides, the equivalent

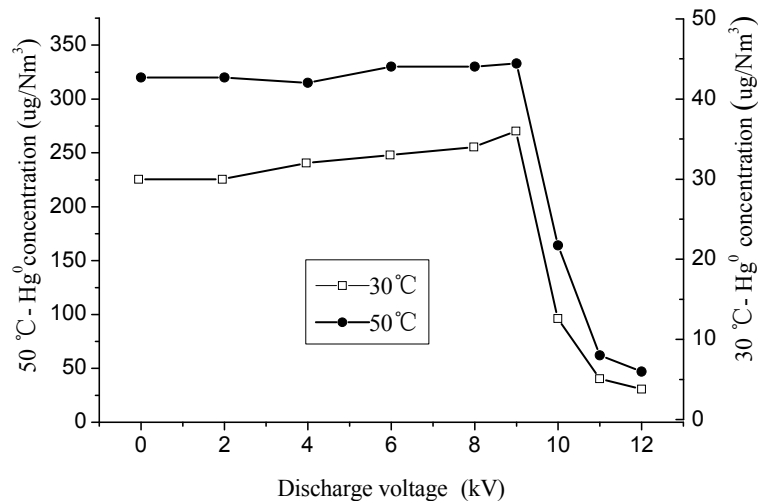
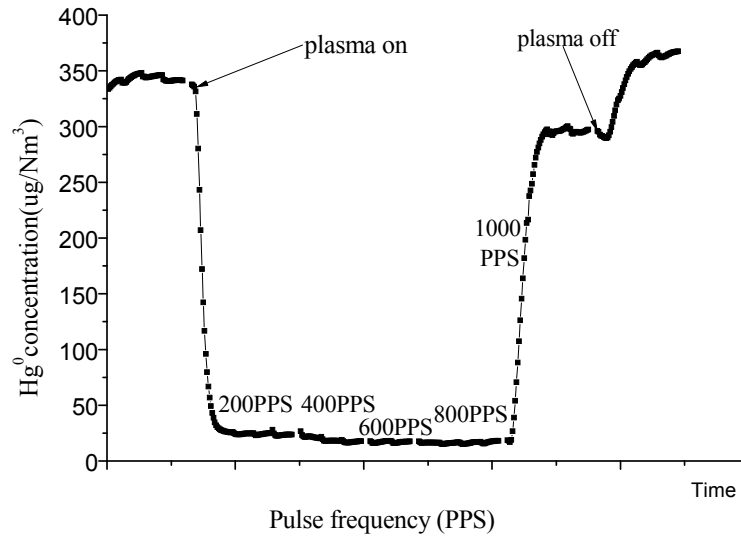


Fig. 2 Variation of Hg<sup>0</sup> concentration with different discharge voltage (O<sub>2</sub>) (10%).



**Fig. 3** Variation of Hg<sup>0</sup> concentration with different pulse frequency (O<sub>2</sub>) (10%).

capacitance of the dielectric layer and the leak inductance of set-up transformer induced resonance.

### 3.2 Influence of Oxygen

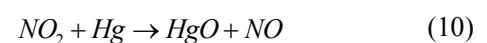
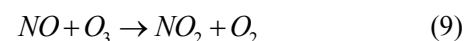
The influence of oxygen concentration on the Hg<sup>0</sup> oxidation was investigated by varying the O<sub>2</sub> concentration from 0% to 20%. Experimental results show that the mercury removal effect is much better than that without O<sub>2</sub> added. With the increase of O<sub>2</sub>, Hg<sup>0</sup> oxidation efficiency increased slowly. Under the condition of 20% O<sub>2</sub>, the oxidation efficiency of Hg<sup>0</sup> is 96.75% at 600 PPS. The concentration of O<sub>2</sub> in flue gas is 10% in commonly. When O<sub>2</sub> concentration is 10%, Hg<sup>0</sup> oxidation efficiency can reach 95.14%, and Hg<sup>0</sup> concentration can be reduced to 17.5 μg/Nm<sup>3</sup>, so the optimal concentration is 10% O<sub>2</sub>.

It should be noticed that some Hg<sup>0</sup> was oxidized even at 0% O<sub>2</sub> conditions, as can be found in Fig. 4. The reason may be that even the balance gas is 99.999% high purity nitrogen, there's still some oxygen as impurities. The high purity nitrogen is produced by air-separation method, the impurity should mainly be O<sub>2</sub> and the amount could be around 10 ppm in the N<sub>2</sub> gas [15]. Although, the oxygen concentration is extremely low, it's still substantial relative to the 300 μg/Nm<sup>3</sup> of Hg<sup>0</sup> (about 33 ppb), for the oxygen concentration is 300 times up on the mercury

concentration. When the plasma discharges, trace amounts of O<sub>2</sub> will produce O and O<sub>3</sub>, so as to realize the oxidation of elemental mercury. In addition, the oxidation efficiency is greatly influenced by the pulse frequency, which is the best at 600 PPS.

### 3.3 Influence of NO and SO<sub>2</sub>

NO is the major O, O<sub>3</sub> scavenger compared with Hg<sup>0</sup>, because the reaction rate in constant of reaction Eq. (9) is five orders of magnitude higher than that of reaction Eq. (8), and NO concentration is much higher than mercury concentration, so Hg<sup>0</sup> oxidation decreased with the increasing NO [18]. Therefore, when the NO concentration increased from 0 ppm to 200 ppm, mercury oxidation rate reduced from 94.46% to 21.59%. When gas exists in a certain amount of high NO<sub>x</sub> (e.g. NO<sub>2</sub> and NO<sub>3</sub>), Hg<sup>0</sup> has been oxidized through reaction Eqs. (10) and (11) [19]. Therefore, when the concentration of NO increased from 200 ppm to 300 ppm, the oxidation rate of elemental mercury increased from 21.59% to 29.69%, eccentrically.



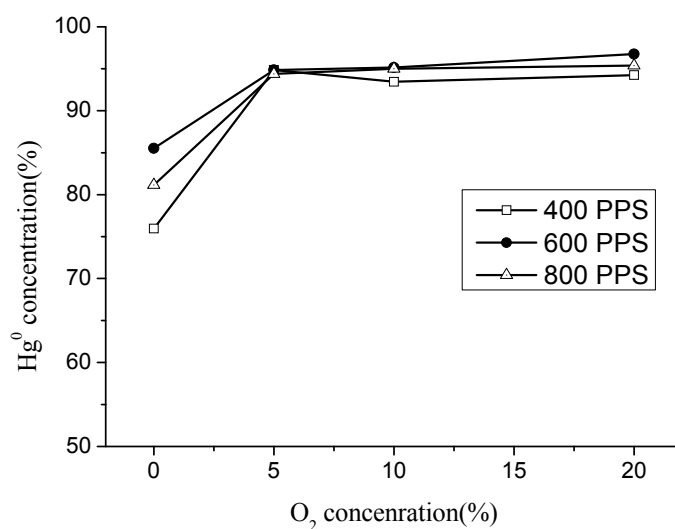


Fig. 4 Effect of O<sub>2</sub> concentration on the oxidation of mercury.

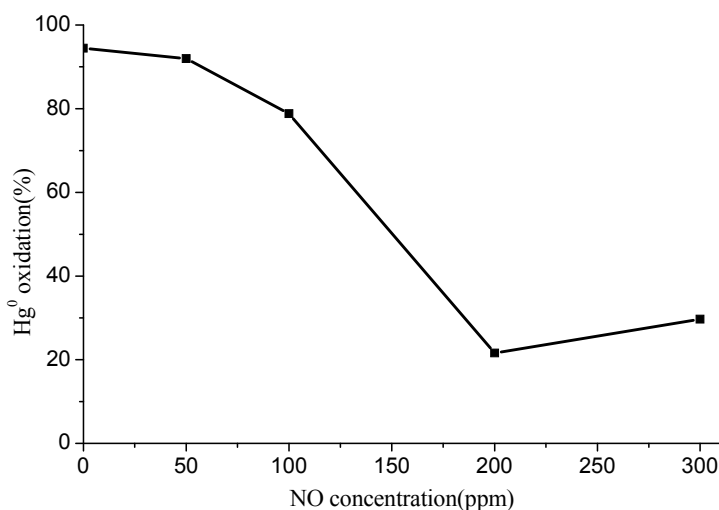
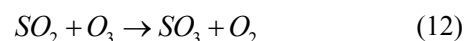


Fig. 5 Effect of NO concentration on the oxidation of mercury (O<sub>2</sub>) (10%).

When the concentration of SO<sub>2</sub> increased from 0 ppm to 500 ppm, mercury oxidation rate reduced from 97.8% to 78.4%. Because with the existence of oxygen, the main active substance is O<sub>3</sub> and O in the process of pulse corona discharge. SO<sub>2</sub> is mainly oxidized by reaction Eq. (12), while, due to the reaction rate is less than that in Eq. (8), so there is little effect on the oxidation of mercury. But SO<sub>2</sub> could react with O with the reaction Eq. (13), so the elemental mercury oxidation rate decreased. At the same time, because of the consumption of HgO with reaction Eq. (14), it will further improve Hg<sup>0</sup> oxidation. There has always been a dispute for the

influence of SO<sub>2</sub> on mercury, and the difference of non-thermal plasma reactor types may be the main reason.



### 3.4 Influence of HCl

In Fig. 7, with the increase of HCl concentration, more Hg<sup>0</sup> is oxidized. The reactor can produce high

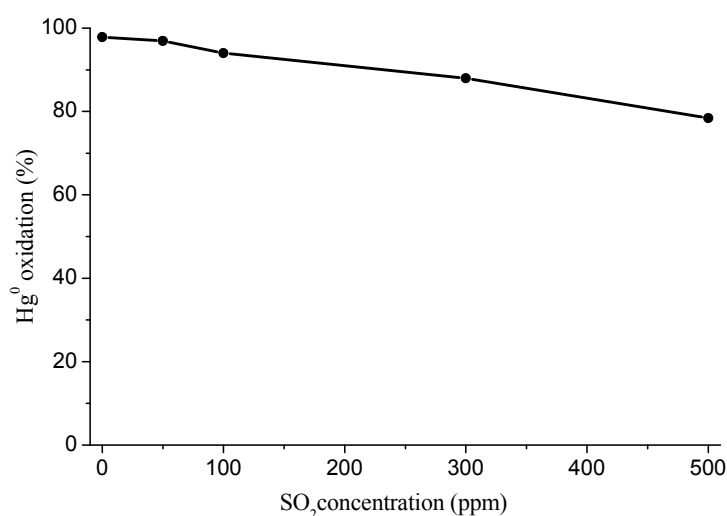


Fig. 6 Effect of  $\text{SO}_2$  concentration on the oxidation of mercury ( $\text{O}_2$ ) (10%).

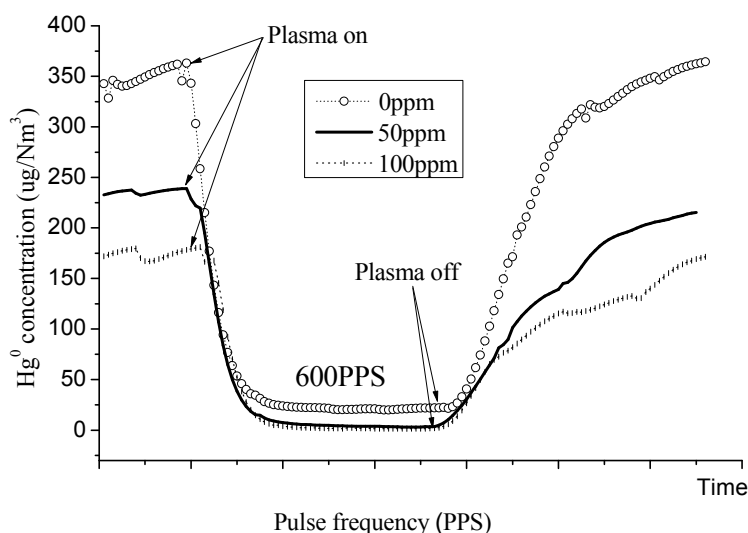
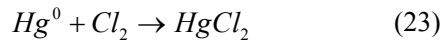
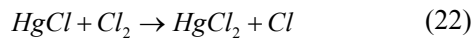
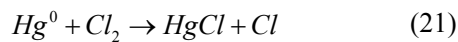
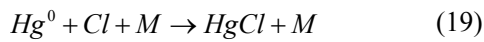
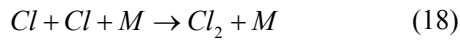
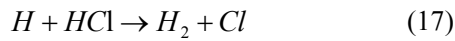
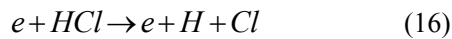
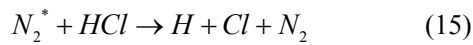


Fig. 7 Effect of  $\text{HCl}$  on oxidation of mercury ( $\text{O}_2$ ) (10%).

energy electron in the pulse discharge process. These electrons undergo collisions with  $\text{N}_2$  buffer gas molecules, which leads to the formation of electronically excited  $\text{N}_2$  molecules ( $\text{N}_2^*$ ), the decomposition of  $\text{N}_2$  into  $\text{N}$  atoms and the production of  $\text{N}_2^*$ . These species, together with energetic electrons, trigger the formation of  $\text{H}$  and  $\text{Cl}$  with the presence of  $\text{HCl}$ , as shown in reaction Eq. (15).  $\text{HCl}$  may also directly be bombarded to decomposition by high energy electron in the process of discharge, as shown in the reaction Eqs. (16) and (17). They can directly oxidize  $\text{Hg}^0$  even at low temperature where

the thermal conversion of  $\text{Hg}^0$  to  $\text{HgCl}_2$  is difficult. Accordingly,  $\text{HCl}$  could play an important role in the formation of oxidants for the oxidation of  $\text{Hg}^0$  in the process of the plasma discharge. And according to Kyung, B. K. [6, 20], emergence of  $\text{HCl}$  can lead to the reaction Eq. (18). According to the experiment, it is reasonable to anticipate that trace amounts of  $\text{Cl}_2$  molecules are likely to be formed by the existence of  $\text{HCl}$ , which is enough to  $\text{Hg}^0$ . Therefore, it is considered that  $\text{Cl}$  atoms and  $\text{Cl}_2$  molecules are responsible for the oxidation of  $\text{Hg}^0$  through the reaction Eqs. (19)-(23):



Focusing on observation of varying concentrations of Hg<sup>0</sup> (0 ppm, 50 ppm and 100 ppm), it's found that with the increase of concentration of HCl, mercury initial concentration decreased significantly which is on the contrary of the mercury oxidation rate. Before the pulse corona discharge, Hg<sup>0</sup> concentration is 357 μg/Nm<sup>3</sup>, 238 μg/Nm<sup>3</sup> and 175 μg/Nm<sup>3</sup>, respectively, and the oxidation rate is 94.4%, 98.7% and 99.2%, correspondingly. So it's concluded that because of the appearance of HCl, a part of Hg<sup>0</sup> have been oxidized before the plasma discharge.

### 3.5 Influence of Moisture

There are always some OH and H atoms generated from H<sub>2</sub>O during the NTP process when moisture existed. In Fig. 8, plasma discharge makes the elemental mercury concentrations decreased obviously. As the moisture concentration is increased from 0.4% to 4%, the Hg<sup>0</sup> oxidation efficiency is decreased from 83.67% to 61.7%. While, when the system only has some residual water vapor, the Hg<sup>0</sup> oxidation efficiency can be as high as 98.13%. Similarly, the moisture concentration in flue gas has an impact on the initial concentration of mercury. When there is 4% moisture, the mercury concentration is approximately 525 μg/Nm<sup>3</sup>, while compared to 0.4% moisture, the mercury concentration is only 245 μg/Nm<sup>3</sup>.

The article made a further study, which set the moisture at 0%, 2%, 3%, 4% and 10%. There is a slight increase of mercury oxidation rate with the increase of concentration of water vapor, then follows a quick drop. Furthermore, OH radical will be generated with the increase of moisture and make a decrease of Hg<sup>0</sup> concentration through reaction Eq. (24). However, the lifetime of HgOH is only 280 μs at 298 K, so that a true kinetic measurement of the recombination reaction would be difficult to achieve in practice [21]. Furthermore, secondary reactions between O<sub>3</sub> and OH will take place and causing consumption and depletion

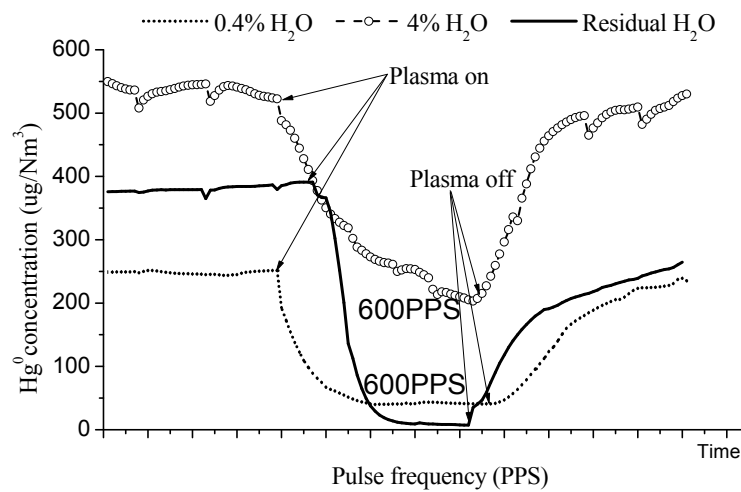


Fig. 8 Effect of H<sub>2</sub>O vapor on oxidation of mercury (O<sub>2</sub>) (10%).

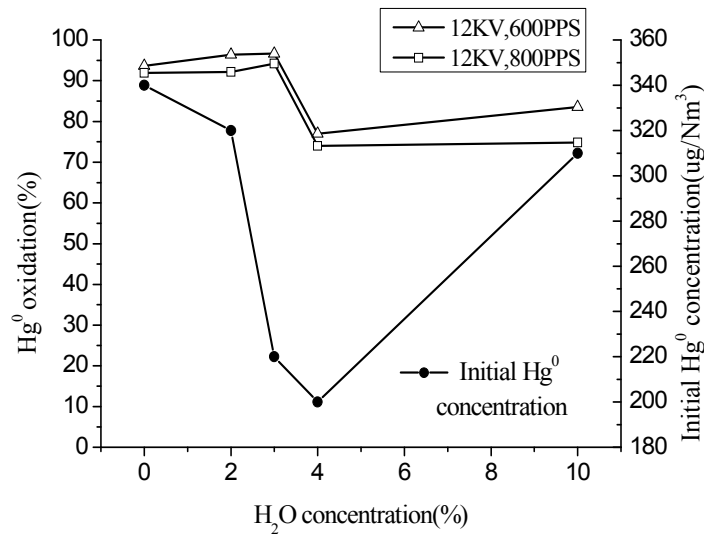


Fig. 9 Effect of H<sub>2</sub>O vapor on oxidation of mercury (O<sub>2</sub>) (10%).

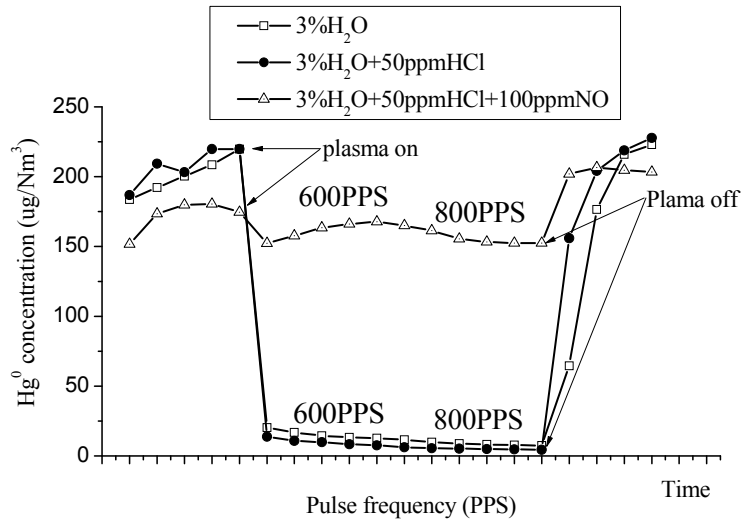
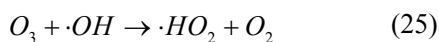


Fig. 10 Effect of the mixed gas components (O<sub>2</sub>) (10%).

of O<sub>3</sub> and O radicals through reaction Eq. (25) [15], and 3% moisture is the best experimental condition. At the same time, it's found that with the increase of moisture, the initial concentration of mercury is not stable, which decreased first and then increased.



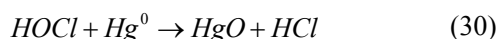
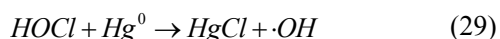
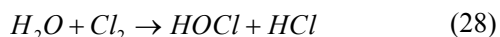
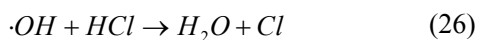
### 3.6 Effect of the Mixed Gas Components

In the actual flue gas, several pollutants exist

simultaneously. To estimate the potential of NTP injection for Hg<sup>0</sup> oxidation in the actual flue gas, the effect of the mixed gas components, including O<sub>2</sub>, H<sub>2</sub>O, HCl and NO was studied. Fig. 10 shows the influence of 50 ppm HCl on the Hg<sup>0</sup> oxidation with 10% O<sub>2</sub> and 3% H<sub>2</sub>O coexisting. OH could react with HCl to generate Cl, and further generating HOCl, involving in the oxidation Hg<sup>0</sup> and increasing of the oxidation of mercury through reaction Eqs. (26)-(30) [16]. Elemental mercury concentration further reduce from 12.7 μg/Nm<sup>3</sup> to 7.7 μg/Nm<sup>3</sup> at 600 PPS pulse frequency. When discharge frequency is 800 PPS,



elemental mercury concentration reduce from 7.4  $\mu\text{g}/\text{Nm}^3$  to 4.5  $\mu\text{g}/\text{Nm}^3$ , and Hg<sup>0</sup> oxidation rate is up to 97.95%.



While, the oxidation efficiency obviously decreased with 100 ppm NO added in this mixed system of 10% O<sub>2</sub>/3% H<sub>2</sub>O/50 ppm HCl. Hg<sup>0</sup> concentration could be down to 152.5  $\mu\text{g}/\text{Nm}^3$  and the oxidation rate reduces to 20% at 800 PPS. So, in the process of mercury on the plasma technology application, NO concentration in the flue gas must be strictly controlled.

#### 4. Conclusion

In this study, the effects of discharge voltage, pulse frequency, concentration of gases such as O<sub>2</sub>, SO<sub>2</sub>, NO, HCl and H<sub>2</sub>O on Hg<sup>0</sup> oxidation in simulated flue gas were systematically investigated and the following conclusions were obtained:

(1) When the discharge voltage is higher than 10 kV, there is a sharp decrease of Hg<sup>0</sup> concentration which is decreased from 333  $\mu\text{g}/\text{Nm}^3$  to 47  $\mu\text{g}/\text{Nm}^3$  and from 36  $\mu\text{g}/\text{Nm}^3$  to 3.8  $\mu\text{g}/\text{Nm}^3$ , respectively. Onset voltage for pulse corona induced plasma is about 10 kV. With the increase of pulse frequency, the Hg<sup>0</sup> concentration falls rapidly at first but then rises rapidly, and 600-800 PPS is the best choice;

(2) Mercury removal efficiency is much better than that without O<sub>2</sub> added. With the increase of O<sub>2</sub> concentration, Hg<sup>0</sup> oxidation efficiency increases. When the O<sub>2</sub> concentration is 10%, Hg<sup>0</sup> oxidation efficiency can reach 95.14%, and Hg<sup>0</sup> concentration can decrease to 17.5  $\mu\text{g}/\text{Nm}^3$ ;

(3) NO is the major O, O<sub>3</sub> scavenger compared with

Hg<sup>0</sup>. When NO concentration increased from 0 ppm to 200 ppm, mercury oxidation rate reduced from 94.46% to 21.59%. While, when SO<sub>2</sub> concentration increased from 0 ppm to 500 ppm, mercury oxidation rate only decreased from 97.8% to 78.4%;

(4) With the increase of HCl concentration, more Hg<sup>0</sup> were oxidized. When moisture was set to 0%, 2%, 3%, 4% and 10%, there's a slight increase of mercury oxidation with the increase of water vapor concentration, then follows a quick drop, and 3% moisture is the best experimental condition;

(5) In this mixed gas components, with 10% O<sub>2</sub>/3% H<sub>2</sub>O/50 ppm HCl coexisted, Hg<sup>0</sup> oxidation rate is up to 97.95% at 800 PPS pulse frequency. While, the oxidation efficiency obviously decreased with 100 ppm NO added. Hg<sup>0</sup> concentration only decreased to 152.5  $\mu\text{g}/\text{Nm}^3$  and the oxidation rate is only 20% at 800 PPS.

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