Oxidation of elemental mercury with non-thermal plasma coupled with a wet process

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HIGHLIGHTS

- Method of dielectric barrier discharge coupled with wet process for Hg^0 oxidation was developed.
- Compared with the DBD system, the Hg oxidation efficiency increased after adding wet process.
- High temperatures promote the oxidation of Hg^0 in the DBD system.
- NO inhibits Hg^0 oxidation, while SO_2 has positive impact on Hg^0 oxidation in wet-DBD system.

ARTICLE INFO

ABSTRACT

The oxidation of the elemental mercury (Hg^0) by non-thermal plasma (NTP) coupled with a wet process has been studied. Effects of oxygen content, supplied voltage, gas temperature, wet process and NO/SO_2 concentration on Hg^0 oxidation were investigated using a dielectric barrier discharge reactor. Results indicate that both O and O_3 generated by O_2 during the discharge process contribute to the oxidation of Hg^0. The increase of O_2 content improves the Hg oxidation efficiency. It was noted that the Hg oxidation efficiency declines when the supplied voltage is too high, there was an optimal voltage level for Hg^0 oxidation. High temperatures promote oxidation of Hg^0. The main reason for this is that the high temperature increases the generation of O atoms and the reaction rate of O_3 with Hg^0. Compared with the NTP process only, the Hg oxidation efficiency increases from 43 to 82% after adding a wet process when the supplied voltage was 8 kV at room temperature. The presence of NO hindered mercury oxidation due to competitive consumption of O_3. However, at the level of 100 ppm NO, more than 50% Hg oxidation efficiency could be achieved in the NTP combined with wet process system. It is found SO_2 enhance oxidation of Hg^0, because of the positive effect of SO_3 generated in the NTP environment.

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1. Introduction

Mercury is a global circulation pollution element because of its high toxicity, volatility, bioaccumulation and impacts on human health and the ecosystem [1]. Mercury emission from coal-fired power plants is the main source of anthropogenic mercury emissions [2]. Depending on coal combustion conditions, mercury species in the flue gas can be classified into three forms: elemental mercury (Hg^0), oxidized mercury (Hg^{2+}) and particle-associated mercury (Hg_p) [3–5]. Hg^{2+} and Hg_p can be efficiently captured by the conventional air pollution control devices (APCDs) such as electrostatic precipitators (ESPs), wet flue gas desulphurization (WFGD) and wet scrubbers [6–8]. While Hg^0 accounts for a significant portion of the mercury species in the flue gas, it is hard to remove because of its high volatility and low solubility. Therefore converting Hg^0 into Hg^{2+} is an effective way to promote the mercury reduction.

Sorbents injection methods have been widely studied and implemented for Hg^0 control in both laboratory and large-scale applications. Considerable efforts have been made to remove Hg^0 with sorbents in the past years [9–11]. However, some factors hinder the application of mercury absorption technology, including extremely low mercury concentrations, short residence time and

high temperatures [12]. To address this problem, non-thermal plasma (NTP) has been proposed, which can remove NO, SO_2 and trace elements simultaneously [13,14]. Hg^0 can be oxidized by reactive radicals such as O, O_3, OH, etc induced by high energy electrons in NTP. Chen et al. reported that Hg^0 can be simultaneously removed along with NO_x and SO_2, and the presence of NO_x enhances mercury oxidation [12]. Byun and Ko et al. studied the removal mechanism of Hg^0 by using a non-thermal plasma and found that O_3 plays an important role in the oxidation of Hg^0 [15–17]. Yang et al. investigated the effect of non-thermal plasma coupled with photocatalyst on the oxidation of Hg^0 [18]. Wang et al. [19] and Niu et al. [20] researched the influence of other flue gas components such as O_2, H_2O, NO and SO_2 etc. on elemental mercury oxidation by NTP, both studies reported that NO hindered the oxidation of Hg^0. Stergaršek et al. studied the removal of Hg^0 in a WFGD by catalytic oxidation with air, indicating that water has an effect on Hg^0 oxidation [21]. To the knowledge of the authors of this paper, most studies that investigate Hg^0 oxidation by NTP focus on the homogeneous reactions between gaseous active species and Hg^0. However, no systematic research on mercury oxidation by NTP combined with a wet process has been carried out so far. The actual flue gas from power plant includes an environment saturated with water, such as in a WFGD. Therefore, the method of non-thermal plasma coupled with a wet process is likely to make sense for implementation in practical applications.

In this paper, a dielectric barrier discharge (DBD) system was used to oxidize Hg^0. DBD is a method to produce non-thermal plasma at atmospheric pressure, which adapted to a wide range of temperature and pressure conditions [22]. A bubble reactor contained with deionized water was arranged after DBD reactor to investigate the effect of NTP combined with a wet process on mercury oxidation. The synergetic effects due to the presence of O_2, NO and SO_2 were also investigated in this study.

2. Experiment

The experimental system diagram is shown in Fig. 1. It consists of a gas feeding system, a dielectric barrier discharge reactor, a high voltage power supply, a bubble reactor, a mercury analysis system and an exhaust gas treatment system. Simulated flue gas was prepared by direct mixing of N_2, O_2, NO (1.02%, N_2 as balance gas) and SO_2 (1.03%, N_2 as balance gas) in a gas mixing chamber. The flow rate of each gas was accurately controlled independently by individual mass flow controllers (MFC). The total flow rate of the simulated flue gas was measured by flow meter and was controlled at 5 L/min in all experiments. The concentration of NO and SO_2 in the simulated flue gas was measured by a flue gas analyzer (Testo 350). Hg^0 was supplied from the mercury source of a Continuous Emission Monitor system (PSA 10.536 CAVKIT) and the initial concentration of Hg^0 was set at 50 μg/m³. The simulated gas was made to pass through the DBD reactor and the bubble reactor in tandem. Then the outlet concentration of Hg^0 was tested by a mercury analyzer (Lumex RA-915⁺). To monitor the outlet concentration of O_3 produced by DBD, an ozone detector (TD1198) was fitted at the tail-end of the system. A digital oscilloscope was used to obtain Lissajous figures for estimating power consumption in the DBD reactor.

The DBD reactor is a coaxial cylinder type structure with an inner discharge electrode, an quartz tube and an outer electrode. The inner diameter of the quartz tube is 21 mm and the outer diameter of the inner discharge electrode is 15 mm, to make sure there is a 3 mm discharge gap. The thickness of the quartz dielectric is 2 mm. The inner discharge electrode, acting as a ground electrode, is a screw-thread copper rod and the flutes pitch is 1 mm. A copper mesh with a length of 150 mm covering the outer surface of the quartz tube, serves as the active electrode, which is connected to an anode of a high voltage AC power supply.

Since it was difficult to control the temperature of the DBD reactor directly, a gas preheater was set before the reactor to preheat the simulated flue gas to temperatures of 363 K and 403 K, to simulate typical power plant exhaust temperatures. To investigate the effect of adding wet process on Hg^0 oxidization, a bubble reactor (i.d. 50 mm, height 400 mm) filled with 200 mL of deionized water was arranged after the DBD reactor. To keep similar resident times for all tests, the empty bubble reactor was still retained when no wet process was tested in the experiments.

In order to evaluate the effect of the oxidation of elemental mercury under a variety of experimental conditions, a Hg oxidation efficiency was defined as follows:

$$\eta_{Hg} = \frac{C_{Hg,off} - C_{Hg,on}}{C_{Hg,off}} \times 100\% \tag{1}$$

where η_{Hg} is the Hg oxidation efficiency, $C_{Hg,on}$ and $C_{Hg,off}$ are the concentrations of Hg^0 when the DBD was turned on or off, respectively.
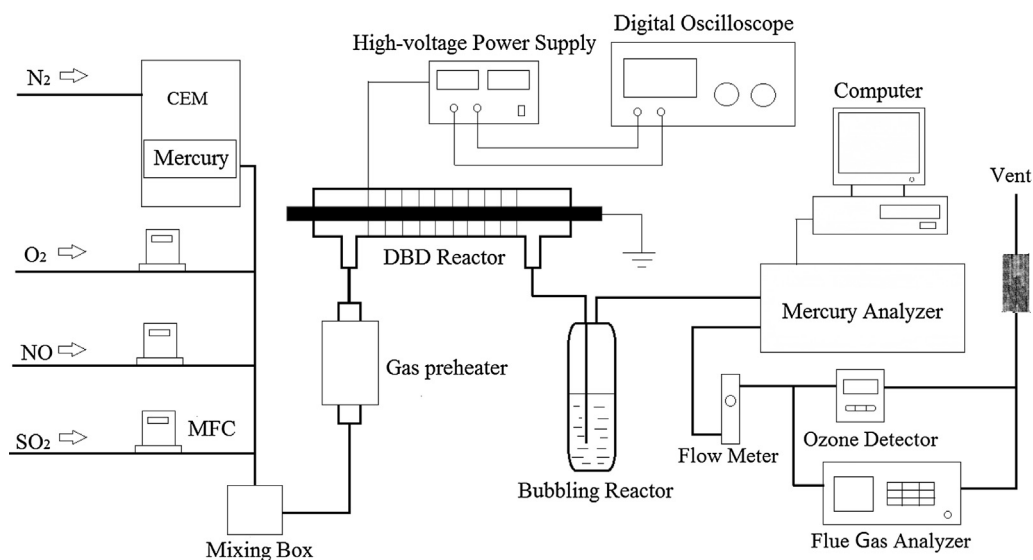

Fig. 1. Experimental apparatus for oxidation of mercury by NTP combined with a wet process.