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A study on removal of elemental mercury in flue gas using fenton solution

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HIGHLIGHTS

- A novel technique on oxidation of Hg^0 using Fenton was proposed.
- The effects of several process parameters on Hg^0 removal were studied.
- Products and • OH in solution were detected.
- Reaction mechanism of Hg^0 removal was studied.
- Simultaneous removal of Hg^0, NO and SO_2 was also studied.

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ABSTRACT

A novel technique on oxidation-separation of elemental mercury (Hg^0) in flue gas using Fenton solution in a bubbling reactor was proposed. The effects of several process parameters (H_2O_2 concentration, Hg^0 inlet concentration, Fe^{2+} concentration, solution temperature, solution pH, gas flow) and several flue gas components (NO, SO_2, O_2, CO_2, inorganic ions and particulate matters on Hg^0 removal were studied. The results indicate that H_2O_2 concentration, Fe^{2+} concentration, solution pH and gas flow have great effects on Hg^0 removal. Solution temperature, Hg^0, NO, SO_2, $CO_3{}^{2-}$ and $HCO_3{}^{-}$ concentrations also have significant effects on Hg^0 removal. However, Cl^-, $SO_4{}^{2-}$, $NO_3{}^-$, O_2 and CO_2 concentrations only have slight effects on Hg^0 removal. Furthermore, reaction mechanism of Hg^0 removal and simultaneous removal process of Hg^0, NO and SO_2 were also studied. Hg^0 is removed by oxidation of •OH and oxidation of H_2O_2. The simultaneous removal efficiencies of 100% for SO_2, 100% for Hg^0 and 88.3% for NO were obtained under optimal test conditions. The results demonstrated the feasibility of Hg^0 removal and simultaneous removal of Hg^0, SO_2 and NO using Fenton solution in a bubbling reactor.

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1. Introduction

Emission of mercury from combustion sources such as coal-fired boilers, industrial furnaces and waste incinerators, which accounts for more than 90% of all anthropogenic mercury emissions, has attracted wide attention due to its increased level in the environment and in the food chain, and bioaccumulation of its compounds [1–3]. Particulate mercury, divalent mercury and elemental mercury are the three most common forms of mercury in typical flue gas. Particulate mercury and divalent mercury can be effectively captured using traditional dusting and washing equipments. However, elemental mercury is very difficult to capture because it has high volatility in room temperature and low solubility in water [1]. Thus developing effective removal techniques of elemental mercury is an important research topic in the field of energy and environment. In recent years, a number of elemental mercury removal technologies have been developed, and according to the removal principle, they may be divided into two categories of adsorption and oxidation [2]. Some adsorbents such as metal oxides, calcium-based materials, carbon-based materials, precious metals and natural mineral materials have been developed to remove elemental mercury [3–6]. In addition, many oxidation techniques and oxidants such as catalytic oxidation, photochemical oxidation, plasma oxidation, ozonation, photocatalytic oxidation, potassium permanganate, sodium chlorite, persulfate and UV/H_2O_2

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oxidation also have been developed to remove elemental mercury [7–16], and have shown good prospects.

Fenton solution can produce hydroxyl free radicals (•OH) with very strong oxidizing potential to degrade various organic pollutants in wastewater and has attracted wide attention duo to its strong oxidizing, simple process and no secondary pollution [17–23]. In the flue gas purification field, Guo et al. and Liu et al. used Fenton solution to remove NO and simultaneously remove NO and SO_2 from flue gas in a bubbling reactor respectively and studied the effects of several parameters on NO and SO_2 removals and the removal products [24,25]. Furthermore, Lu et al. used Fenton-like reactions to remove Hg^0 from flue gas in a simple bubbling reactor, and preliminarily studied the effects of several parameters such as solution pH, residence time, volume of solution on removal of Hg^0 [26]. However, so far, there are few relevant literatures on Hg^0 removal from flue gas by using classic Fenton solution. These developments and problems prompt us to attempt to study the removal of Hg^0 from flue gas using Fenton solution. The main purpose of this article is to study the effects of process parameters such as H_2O_2 concentration, Hg^0 inlet concentration, Fe^{2+} concentration, solution temperature, solution pH, gas flow and flue gas components such as NO, SO_2, O_2, and CO_2, inorganic ions and particulate matters on Hg^0 removal using Fenton solution in a bubbling reactor. Furthermore, the reaction mechanism of Hg^0 removal and the simultaneous removal process of Hg^0, NO and SO_2 were also studied preliminarily. These results will be useful for the further development and industrial applications of this technology.

2. Experimental

2.1. Experimental installation

It can be seen in Fig. 1, the experimental installation mainly consists of a flue gas blending system, an analytical system and a bubbling reactor. Five cylinder gases, including CO_2, NO, O_2, SO_2, and N_2 (Purity, 99.99%, Suzhou Jinhong Specialty Gas Co., Ltd., China), and Hg generator (VICI Metronics, USA) are used to make carrier gas and simulated flue gas containing-$CO_2/NO/O_2/SO_2/N_2/Hg^0$. The bubbling reactor (9.0 cm i.d. and 40 cm length), with a jacket heat exchanger, a bubbler, a reactor cover, a pump and a thermometer, is made of Plexiglass. Reactor cover is used to add solution and bubbler is used to distribute simulated flue gas. Jacket heat exchanger, pump, constant temperature water bath and thermometer are jointly used to control solution temperature. Flue gas analyzer (MRU-VARIO PLUS, Germany) and Hg analyzer (QM201H, Suzhou Qingan Instrument Co., China) are used to measure the concentrations of CO_2, SO_2, NO, O_2 and Hg^0, respectively.

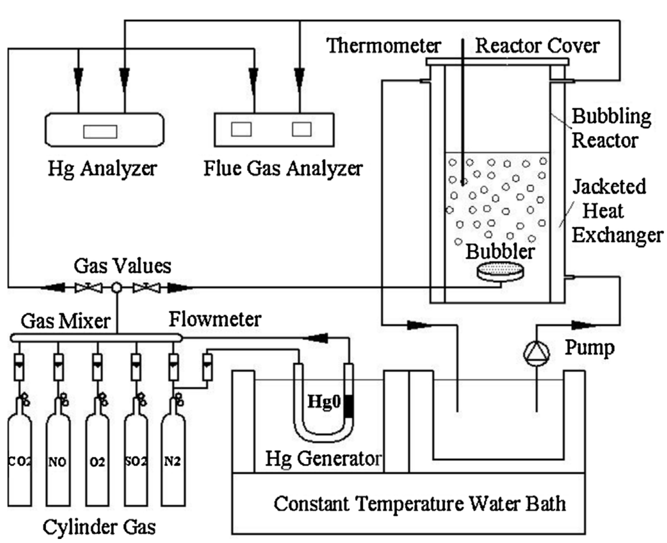

Fig. 1. Schematic diagram of experimental installation.

2.2. Experimental procedures

The Hg^0 analyzer uses a cycle measuring mode of 1 min sampling-3 min measuring -4 min cleaning (gold amalgamation method is used to collect Hg^0 and fluorescence spectroscopy method is used to determine Hg^0 concentration), thus obtaining a measuring value often requires 8 min. The average concentration within 16 min can be obtained by taking the average value of the two instantaneous values in the 8th min and 16th min. 800 mL/min of flue gas was prepared by jointly using the cylinder gases, the Hg generator and the flowmeters. The inlet concentrations of flue gas components such as CO_2, NO, O_2, SO_2 and Hg^0 were detected using the flue gas analyzer and the Hg analyzer. 600 mL of Fenton solution (Fe^{2+}/H_2O_2) was prepared by commercial $FeCl_2{\cdot}6H_2O$ and H_2O_2 reagents (AR, Sinopharm Chemical Reagent Co., Ltd., China) and deionized water. The Fenton solution was added to the bubbling reactor by opening the reactor cover after the solution pH was adjusted using an acidometer. The solution temperature was adjusted to the required values by jointly using the constant temperature water bath, the pump and the thermometer. The flue gas entered the bubbling reactor to make an oxidation–absorption reaction by switching two gas valves. The outlet concentrations of NO, SO_2 and Hg^0 were detected using the flue gas analyzer and the Hg analyzer. Every test time was kept for 16 min, and the average value of two instantaneous concentrations in the 8th min and 16th min was used as the outlet concentration of NO, SO_2, CO_2 and Hg^0. The reaction solutions were analyzed by ion chromatography (Metrohm-861). The key •OH was detected by the MS detector of liquid-mass spectrometry combining with the capture agent salicylic acid (SA). The concentrations of mercury were analyzed using liquid fluorescence mercury analyzer, and the analyzed method referenced in literature [1].

2.3. Removal efficiency

The concentrations of $SO_2/NO/Hg^0$ detected through gas bypass are used as the inlet concentrations. The average value of two instantaneous concentrations of $SO_2/NO/Hg^0$ in the 8th min and 16th min detected through reactor outlet are used as the outlet concentrations. Removal efficiencies of $SO_2/NO/Hg^0$ in flue gas can be calculated using the expression (1):

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

where η is the $SO_2/NO/Hg^0$ removal efficiencies; C_{in} is the inlet concentration of $SO_2/NO/Hg^0$ in flue gas; C_{out} is the outlet concentration of $SO_2/NO/Hg^0$ in flue gas.

3. Results and discussions

3.1. Effects of H_2O_2 concentration

Fig. 2 shows the effects of H_2O_2 concentration on Hg^0 removal efficiency under different Fe^{2+} concentrations. The results show that with the increase from 0 to 1.2 mol/L of H_2O_2 concentration, Hg^0 removal efficiency sharply increases from 0 to 91.2% under 0.008 mol/L of Fe^{2+} concentration, and sharply increases from 0 to 43.1% under 0.002 mol/L of Fe^{2+} concentration, respectively. The

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