



# Elemental mercury removal from flue gas using heat and $\text{Co}^{2+}/\text{Fe}^{2+}$ coactivated oxone oxidation system

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## HIGHLIGHTS

- Novel  $\text{Hg}^0$  removal process using heat and  $\text{Co}^{2+}/\text{Fe}^{2+}$  coactivated Oxone is developed.
- Active species, products and mechanism of  $\text{Hg}^0$  removal process are investigated.
- Mass transfer-reaction kinetic law of  $\text{Hg}^0$  removal process is also revealed.
- Heat and  $\text{Co}^{2+}/\text{Fe}^{2+}$  have a very significant synergistic effect.
- $\text{Hg}^0$  removal processes belong to fast reactions under optimized conditions.

## ARTICLE INFO

### Keywords:

Heat and  $\text{Co}^{2+}/\text{Fe}^{2+}$  coactivated oxone  
Elemental mercury ( $\text{Hg}^0$ )  
 $\cdot\text{OH}$  radical  
 $\text{SO}_4^{\cdot-}$  radical

## ABSTRACT

Oxidation removal process of elemental mercury ( $\text{Hg}^0$ ) from flue gas using heat and  $\text{Co}^{2+}/\text{Fe}^{2+}$  coactivated Oxone oxidation system in a spraying reactor was developed. The effects of several influencing factors and main flue gas compositions on  $\text{Hg}^0$  removal, and the active species, products, mechanism and mass transfer-reaction kinetic law of  $\text{Hg}^0$  removal process were investigated. The results show that heat and  $\text{Co}^{2+}/\text{Fe}^{2+}$  have a significant synergistic effect for activating Oxone to produce free radicals and promote  $\text{Hg}^0$  removal.  $\text{Hg}^0$  removal can be improved via increasing Oxone concentration,  $\text{Co}^{2+}/\text{Fe}^{2+}$  concentration, activation temperature or  $\text{O}_2$  concentration, and is inhibited with increasing solution pH,  $\text{Hg}^0$  inlet concentration or NO concentration. Changing  $\text{SO}_2$  concentration have double impacts on  $\text{Hg}^0$  removal.  $\text{SO}_4^{\cdot-}$  and  $\cdot\text{OH}$  produced from coactivation of heat and  $\text{Co}^{2+}/\text{Fe}^{2+}$  are the key oxidants, which play a leading role in  $\text{Hg}^0$  removal. Oxone plays a complementary role in  $\text{Hg}^0$  removal.  $\text{Hg}^0$  removal processes using heat/ $\text{Co}^{2+}$  and heat/ $\text{Fe}^{2+}$  coactivated Oxone systems belong to fast reactions under optimized experimental conditions, and meets a total 2.8-order reaction (i.e. 1.0-order for  $\text{Hg}^0$ , 1.1-order for Oxone and 0.7-order for  $\text{Co}^{2+}$ ) in  $\text{Co}^{2+}/\text{Oxone}$  activation system, and a total 3.0-order reaction (i.e. 1.0-order for  $\text{Hg}^0$ , 1.4-order for Oxone and 0.6-order for  $\text{Fe}^{2+}$ ) in  $\text{Fe}^{2+}/\text{Oxone}$  activation system. Finally, the key mass transfer and kinetic parameters of  $\text{Hg}^0$  removal processes were also determined.

## 1. Introduction

Emissions of mercury have caused worldwide attention because of its negative influence on human health and ecological environment [1,2]. The mercury from typical coal-fired flue gas mainly consists of particulate bounded mercury ( $\text{Hg}^{\text{P}}$ ), elemental mercury ( $\text{Hg}^0$ ) and oxidized mercury ( $\text{Hg}^{2+}$ ) [3–5]. The  $\text{Hg}^0$  is recognized to be the hardest to be captured owing to its low water solubility [6,7]. To decrease  $\text{Hg}^0$  emission, a great deal of related technologies have been developed, including catalytic oxidation [2,3,7,8], adsorption removal [4,5], oxidation removal, etc [6,9,10]. Based on different oxidants and oxidation principles,  $\text{Hg}^0$  oxidation removal technologies can be further divided

into conventional oxidation and free radical advanced oxidation [6].

Free radical advanced oxidation technologies for  $\text{Hg}^0$  removal mainly include plasma removal [11], photocatalytic oxidation [12], photochemical removal [13,14,15], heat-activated oxidants removal [6], metal oxide-activated oxidants removal [10,16], ion-activated oxidants removal, etc [7,18]. Plasma removal, photocatalytic oxidation and photochemical removal technologies can achieve high  $\text{Hg}^0$  removal efficiency, but they have some disadvantages in investment and operating costs, and system reliability [13,17,18]. Heat and metal oxide-activated oxidants removal technologies have low investment and operating costs, but they usually have a low  $\text{Hg}^0$  removal capacity because of low radical activation efficiency and high mass transfer resistance

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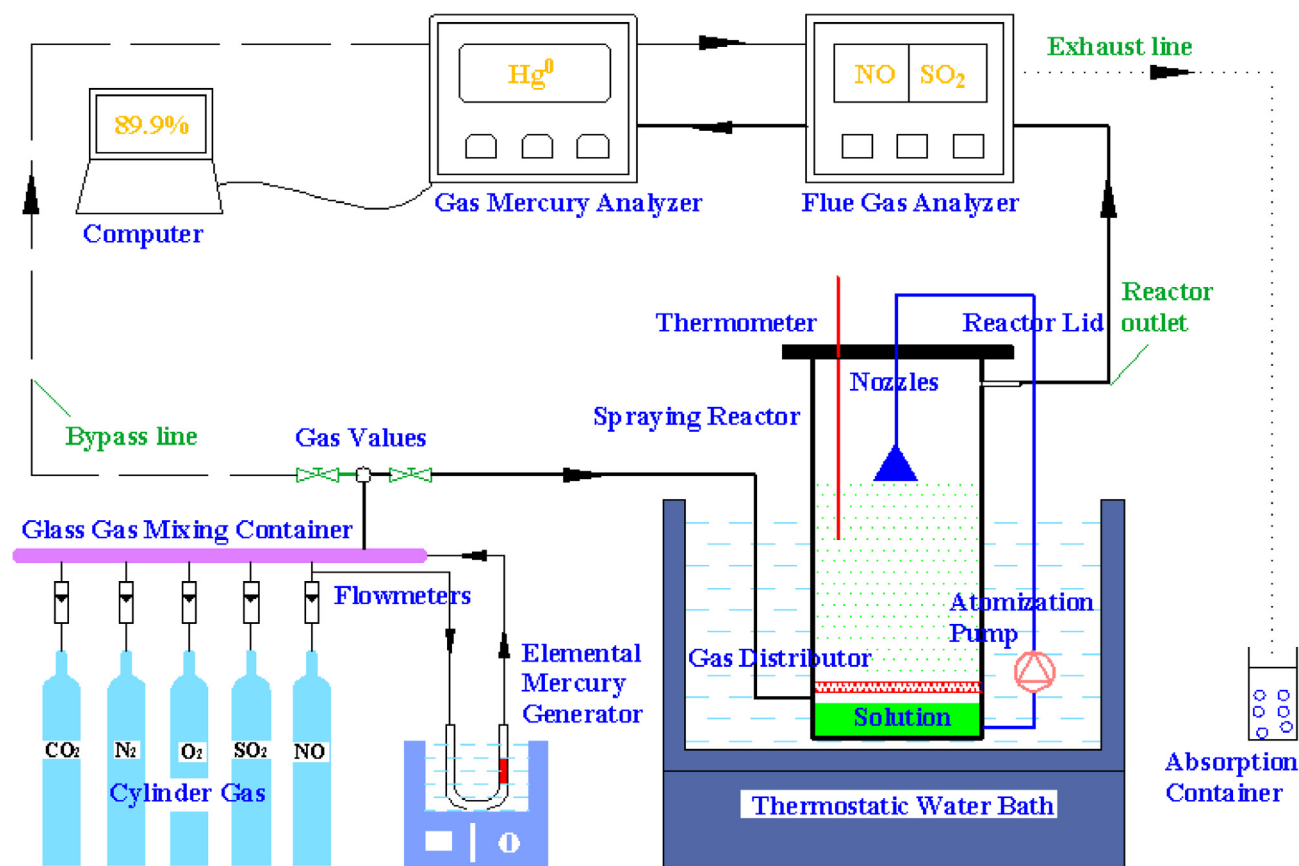


Fig. 1. The schematic diagram of elemental mercury removal system.

[13,17,18]. In comparison, ion-activated oxidants removal technologies have low investment and operating costs, and relatively high  $\text{Hg}^0$  removal efficiency, which has shown good prospects [17,18].

In the previous works [17,18], Fenton reagent ( $\text{Fe}^{2+}/\text{H}_2\text{O}_2$ ) and Fenton-like reagents ( $\text{Fe}^{3+}/\text{H}_2\text{O}_2$  and  $\text{Cu}^{2+}/\text{H}_2\text{O}_2$ ) were used to oxidize  $\text{Hg}^0$  from flue gas, and showed good  $\text{Hg}^0$  removal performance. Nevertheless, Fenton and Fenton-like reagents also showed several obvious disadvantages as follows [17,18]: (1) the reaction between ions and  $\text{H}_2\text{O}_2$  is very violent and uncontrollable, which results in a large self-consumption in ions and  $\text{H}_2\text{O}_2$ ; (2) Fenton and Fenton-like reagents often have a very narrow optimized pH window (the optimum pH is usually between 3 and 5). However, there are a lot of nitrogen oxides, sulfur dioxide and hydrogen halide in the actual coal-fired flue gas, they will become acid after being oxidized or washed. As the reaction progresses, the pH value of the solution will become extremely low, which is not suitable for Fenton and Fenton-like reactions. As shown in Fig. 2(c), the results in this study show that both  $\text{Co}^{2+}/\text{Oxone}$  and  $\text{Fe}^{2+}/\text{Oxone}$  activation systems have a wide range of optimized pH windows, which have a much stronger adaptability in future industrial operations; (3) it is well known that common industrial grade hydrogen peroxide solution contains 72.5% of water. The reaction products will contain very dilute sulfuric acid and nitric acid mixed solutions (because adding hydrogen peroxide will inevitably bring in a lot of water at the same time). This will lead to huge costs and energy consumption in concentrating the reaction products, which hinder the application of this technology.

Oxone is a water-soluble, environmentally friendly, safe to handle and stable oxidant. Especially as a solid oxidant, Oxone has a big advantage in concentrating the reaction products (when solid oxidizer is added, no water is carried). Besides, it is also much easier to store and transport, with a higher security, than commercial  $\text{H}_2\text{O}_2$  solution [19–22]. In the past several years, Oxone-based advanced oxidation

technologies have been widely studied in water treatment field [22]. In the field of flue gas purification, Adewuyi et al. developed the oxidation process of nitric oxide via aqueous solutions of Oxone in a bubble column reactor, and studied the several operating parameters and nitric oxide absorption rates [19]. Recently, the authors reported the removals of NO and  $\text{SO}_2$  using aqueous Oxone with independent or synergistic activation of transition metal ions, heat, UV light and ultrasound in a variety of gas-liquid reactors [20,21,24–25]. Among these methods, transition metal ions activation technologies show excellent development potential for NO and  $\text{SO}_2$  removals because of simple process/device and low costs [20,23]. In this article, our aim is to further explore the  $\text{Hg}^0$  removal using transition metal ions-activated Oxone advanced oxidation technologies. The ultimate goal is to achieve the simultaneous removal of NO,  $\text{SO}_2$  and  $\text{Hg}^0$  from flue gas in a reactor (presently, the simultaneous removal of multi-pollutants from flue gas is a hot issue in the field of air pollution control) [24,26].

In several common transition metal ions such as  $\text{Co}^{2+}$ ,  $\text{Cu}^{2+}$ ,  $\text{Fe}^{3+}$ ,  $\text{Fe}^{2+}$ ,  $\text{Mn}^{2+}$ , etc.  $\text{Co}^{2+}$  has been proved to have the highest free radical activation efficiency for Oxone [22,26–28]. In addition, ferrous sulfate ( $\text{Fe}^{2+}$ ) is the by-product of titanium dioxide production process (in China, the production of titanium dioxide mainly uses sulfuric acid method. Production of one ton of titanium dioxide often will produce 3–4 ton of waste  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ ). According to the statistics, the total yield of the waste  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$  has been more than 5 million tons per year in China [39]. Due to the huge production and presence of high impurities, most of the waste  $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$  can not be used effectively, which even causes environmental pollution [29]. Based on this situation, both of  $\text{Co}^{2+}$  and  $\text{Fe}^{2+}$  will be chosen as the activators of Oxone in this study. In addition, in the previous works [20,21,24,25], the impinging stream reactor and the bubbling reactor were used as the scrubbers. However, as we all know, spraying reactor is the most widely used wet scrubbers in the area of flue gas purification owing to its very

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