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# Absorption characteristics of elemental mercury in mercury chloride solutions

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## ARTICLE INFO

### Article history:

Received 20 December 2013

Revised 18 March 2014

Accepted 28 March 2014

Available online 20 September 2014

### Keywords:

Nonferrous metal

Smelting flue gas

Elemental mercury

Mercury chloride

## ABSTRACT

Elemental mercury ( $\text{Hg}^0$ ) in flue gases can be efficiently captured by mercury chloride ( $\text{HgCl}_2$ ) solution. However, the absorption behaviors and the influencing effects are still poorly understood. The mechanism of  $\text{Hg}^0$  absorption by  $\text{HgCl}_2$  and the factors that control the removal were studied in this paper. It was found that when the mole ratio of  $\text{Cl}^-$  to  $\text{HgCl}_2$  is 10:1, the  $\text{Hg}^0$  removal efficiency is the highest. Among the main mercury chloride species,  $\text{HgCl}_3^-$  is the most efficient ion for  $\text{Hg}^0$  removal in the  $\text{HgCl}_2$  absorption system when moderate concentrations of chloride ions exist. The  $\text{Hg}^0$  absorption reactions in the aqueous phase were investigated computationally using Moller–Plesset perturbation theory. The calculated Gibbs free energies and energy barriers are in excellent agreement with the results obtained from experiments. In the presence of  $\text{SO}_3^{2-}$  and  $\text{SO}_2$ ,  $\text{Hg}^{2+}$  reduction occurred and  $\text{Hg}^0$  removal efficiency decreased. The reduced  $\text{Hg}^0$  removal can be controlled through increased chloride concentration to some degree. Low pH value in  $\text{HgCl}_2$  solution enhanced the  $\text{Hg}^0$  removal efficiency, and the effect was more significant in dilute  $\text{HgCl}_2$  solutions. The presence of  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  did not affect  $\text{Hg}^0$  removal by  $\text{HgCl}_2$ .

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## Introduction

Mercury has attracted global attention due to its high toxicity and persistence in the environment (Barbosa et al., 2001; Winfrey and Rudd, 1990). The “Minamata Convention on Mercury” was recently signed in October, 2013 in Kumamoto, Japan. It is an international treaty aimed to reduce mercury emission globally.

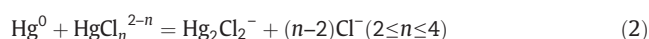
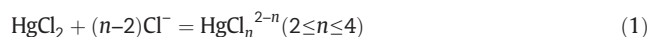
Among the anthropogenic sources of mercury emission, nonferrous metal production, such as the smelting of Pb and Zn sulfide ores, has been recognized as one of the primary sources (UNEP, 2010; Hylander and Herbert, 2008; Li et al., 2010). In China, over 40% of the total mercury emission is from nonferrous metal smelting (Wu et al., 2006; Wang et al., 2006; Pacyna et al., 2010). Therefore, it is urgent to look for an approach to reduce Hg emission in this industry.

As one of the main mercury species in nonferrous metal smelting flue gas, elemental mercury ( $\text{Hg}^0$ ) is hard to remove because of its high volatility and insolubility (Pacyna et al., 2001; Pacyna and Pacyna, 2002). Generally, the major technique for controlling  $\text{Hg}^0$  emission from flue gas is to convert  $\text{Hg}^0$  to its particulate form ( $\text{Hg}^p$ ) or oxidized form ( $\text{Hg}^{2+}$ ) and remove them using existing air pollution control devices, such as electrostatic precipitators or wet flue gas desulfurization units (Qu et al., 2009; Yan et al., 2009; Liu et al., 2011a). However, the concentration of  $\text{Hg}^0$  in nonferrous metal smelting flue gas ranges from several to tens of  $\text{mg}/\text{m}^3$  (Dong, 1994; Wang et al., 2010). It is difficult to remove such high concentrations of  $\text{Hg}^0$  in this industry. Furthermore,  $\text{Hg}^0$  is a valuable resource in many fields such as dentistry, mercury thermometers and gold mining. Therefore, reclaiming mercury from nonferrous metal smelting flue gas is a better choice than

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just removing it by using air pollution control devices for mercury emission control. The mercury chloride absorption technique is an effective  $\text{Hg}^0$  reclamation technology and has been applied in many nonferrous metal smelters.

According to the typical smelting process of nonferrous metal ores, the flue gas from a roasting furnace goes through an electrostatic precipitator, wet scrubber and electrostatic demister to remove particulate matter and clean the flue gas, respectively. Meanwhile, the flue gas is allowed to cool down to about 313 K. Then, the  $\text{Hg}^0$  in the flue gas is reclaimed by mercury chloride absorption technology (Hylander and Herbert, 2008; Wang et al., 2010). The mercury chloride absorption process is based on the oxidation of  $\text{Hg}^0$  by mercuric chloride to form insoluble calomel ( $\text{Hg}_2\text{Cl}_2$ ). The main chemical reactions in the absorption process are summarized as follows (Hylander and Herbert, 2008).



Although the  $\text{Hg}^0$  removal efficiency of the mercury chloride absorption process is about 90%, the outlet  $\text{Hg}^0$  concentration of the mercury chloride absorption process is still very high, and cannot satisfy the discharge standard (Wang et al., 2010). Furthermore, the nonferrous metal smelting flue gas contains high concentrations of  $\text{SO}_2$ , which if absorbed by the mercury chloride absorption solution will reduce the  $\text{Hg}^0$  removal efficiency because  $\text{SO}_3^{2-}$  reduces  $\text{Hg}^{2+}$  in the absorption solution (Liu et al., 2011b). With the emission regulation of  $\text{Hg}^0$  in the nonferrous metal industry becoming increasingly strict, the mercury chloride absorption technology has difficulty meeting the stringent requirements of mercury emission control. Thus, it is important and necessary to improve the mercury chloride absorption technique and enhance its mercury removal performance.

However, relevant reports on the mechanism and main factors affecting mercury chloride absorption technology are lacking, thus limiting its development. This paper

systematically researched the mercury chloride absorption technique and developed a method to enhance it.

## 1. Materials and methods

### 1.1. Experimental apparatus

A typical absorption reactor was applied in this research. As shown in Fig. 1, several cylinder gases were used to provide the simulated flue gas at a flow of 1.0 L/min. Mercury vapor was generated by a mercury bottle in a water bath and was carried by  $\text{N}_2$  gas, and the simulated flue gas passed through a three-neck flask which was filled with 30 mL  $\text{HgCl}_2$  absorption solution. Meanwhile, a pulse reaction system was used to investigate the instantaneous reaction in the absorption solution. Mercury vapor was also generated by a mercury bottle in a water bath and was carried by the simulated flue gas and passed through a three-neck flask. Then, a certain amount of  $\text{Na}_2\text{SO}_3$  solution was injected into the three-neck flask that was filled with 30 mL  $\text{HgCl}_2$  solution to investigate the reduction of  $\text{HgCl}_2$  by  $\text{SO}_3^{2-}$ .  $\text{SO}_2$  was introduced into the reaction system to study the effect of  $\text{SO}_2$  on  $\text{Hg}^0$  removal by  $\text{HgCl}_2$  solution. The average temperature of the simulated flue gas and absorption solution was about 303 and 298 K, respectively. The time for the gas mixture to pass through the absorption solution in the reactor was about 1.2 sec. The initial concentration of  $\text{Hg}^0$  in the inlet gas of the absorption reactor was maintained at about 1.2–2.1  $\text{mg}/\text{m}^3$ , which could be controlled by adjusting the temperature of the water bath and the flow rate of  $\text{N}_2$ . The inlet and outlet  $\text{Hg}^0$  concentrations of the absorption reactor were monitored by a mercury analyzer (SG-921, Jiangfen Ltd., Taizhou, Jiangsu, China). The signal was collected and recorded by a data transition and acquisition device (N2000, Zhida Ltd., Hangzhou, Zhejiang, China). The concentration of  $\text{Hg}^0$  in the flue gas was calibrated by a Lumex mercury analyzer (RA915, Lumex Ltd., St. Petersburg, Russia).

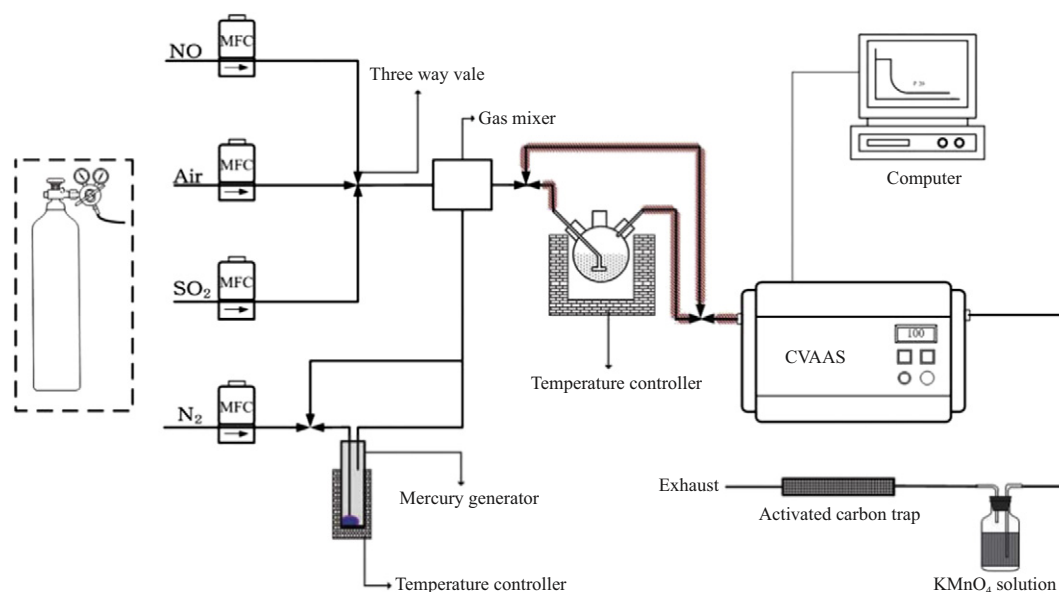


Fig. 1 – Schematic diagram of the absorption experimental apparatus.

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