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Retention of arsenic in coal combustion flue gas at high temperature in the presence of CaO

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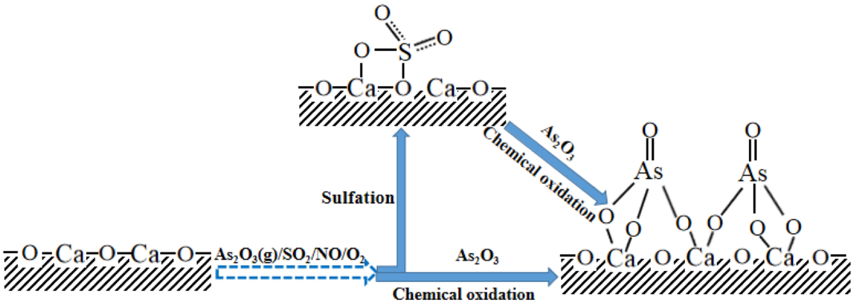
GRAPHICAL ABSTRACT



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ABSTRACT

Numerous studies have demonstrated that the in-situ capture of arsenic by CaO is a promising approach to control arsenic emission from coal-fired flue gas at low and medium temperature. However, the actual temperature of flue gas in furnace exceeds 1000 °C and the reaction mechanism regarding capture of arsenic by CaO at such high temperatures is still unclear. Therefore, in the study, the interaction between arsenic in the vapor phase (As_2O_3(g)) and CaO in the simulated flue gas from 1000 to 1300 °C were investigated. The results indicated that the amount of arsenic captured by CaO increased with the increasing temperature and As_2O_3(g) was fixed into the products in the form of $Ca_3(AsO_4)_2$. Due to the formation of $CaSO_4$ in flue gas, SO_2 can significantly promote the adsorption of arsenic between 1000 °C and 1200 °C. However, with the increase of temperature, this promotion effect gradually faded or even disappeared. On the other hand, in the presence of NO, the slight inhibiting effect on arsenic capture occurred over almost the entire operating temperature range. It was also found that lattice oxygen could also be served as oxidant for the chemical oxidation of As_2O_3(g) in the absence of O_2. The results in this study suggested that the retention of arsenic from coal combustion flue gas in furnace could be facilitated by increasing temperature as well as O_2 concentration and usage amount of high-sulfur coal appropriately.

1. Introduction

In recent years, arsenic has received increasing attention due to its high toxicity and effumability as well as bioaccumulation. Although arsenic in coal is a trace element [1], considering the tremendous coal consumption in China, the total amount of gaseous arsenic discharged

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into environment cannot be underestimated. Therefore, it is imperative to develop arsenic capture technologies that can be exploited to control the arsenic emission of coal-fired power plants.

Gaseous arsenic compounds (such as As_2O_3) derived from coal combustion are converted into larger solid particles by homogeneous or heterogeneous condensation when the temperature drops below 200 °C [2–4]. These arsenic-containing solid particles can be captured through electrostatic precipitator, cyclone dust collector, fabric filer and other dedusting equipments. Nevertheless, at higher temperature, arsenic exists in gaseous form or is concentrated in fine particles after volatilization, which is difficult to remove with conventional dust-cleaning apparatus. Theoretically, As in the flue gas can be condensed by reducing the temperature and then removed by adsorption with activated carbons or other solid adsorbents [5,6], but it causes flowing gas to lose its levitation force. In subsequent operations, the gas needs to be reheated to flow, which undoubtedly increases running costs. Some researches demonstrated that the retention of As_2O_3(g) in coal-fired flue gas at higher temperature can be achieved by injecting mineral materials into coal-fired boilers. During this process, mineral materials can provide an attachable surface for physical adsorption of arsenic [7–9], Meanwhile, arsenous compounds in the vapor phase can be easily transformed into stabilized arsenate on their active sites via chemical oxidation reaction. Consequently, it is a promising approach to reduce emissions of arsenic from coal-fired flue gas [10,11].

Due to the low cost and availability, calcium-based mineral materials have been frequently employed as solid adsorbents to suppress the release of arsenic in coal and control concentration of arsenic in flue gas. Hydrated lime, limestone, kaolinite and bauxite were applied to capture arsenic by Gullett et al. [12]. The results displayed that hydrated lime and limestone exhibited higher capture efficiency for arsenic compared to kaolinite and bauxite. Sterling and Helbe [13] also demonstrated that calcium oxide could efficiently capture gaseous arsenic. Moreover, the interaction between arsenic and CaO has also been lucubrated. Hu et al. [14] studied the retention of arsenic by CaO. The results showed that the process of arsenic capture was a complex combination of physical adsorption and chemical oxidation in lower temperature range. As the operating temperature increased, the chemical processes gradually became the only role in the capture process.

Temperature is a crucial factor affecting the retention of gaseous arsenic by solid adsorbents [15]. Previous researches have demonstrated that the capture process is rather complicated and various arsenate products are formed at different temperatures. Experiments performed by Hu [14] indicated that $Ca_3(AsO_4)_2$ with different crystal structures were produced by reaction between CaO and arsenic in the vapor phase from 300 °C to 1050 °C. Based on the study conducted by Jadhav and Fan [16], the reaction products were both $Ca_3(AsO_4)_2$ at 500 °C and 1000 °C, while gaseous arsenic compounds were detained in CaO in the form of $Ca_2As_2O_7$ in the range of 700–900 °C. Moreover, various reaction trends occurred along with temperature change. Li and Tong [17] investigated the interaction between CaO and As_2O_3(g) from 400 °C to 1000 °C. The results indicated that the capture rate of As_2O_3 by CaO persistently increased with the increase of temperature. However, Jadhav et al. [16] came up with a different conclusion that the retention of arsenic would recede over the temperature range 600–1000 °C, which was mainly attributed to the decomposition of unstable arsenate products and re-release of arsenic oxide at higher temperature. Hu et al [14] supported that increasing the temperature from 300 °C to 750 °C facilitated retention of arsenic while less arsenic was adsorbed above 900 °C.

To date, many researches regarding the capture of arsenic by CaO have been reported and the most of them focus primarily on low and intermediate temperature regions. However, the actual temperature of coal-fired flue gas in furnace usually exceeds 1000 °C and the retention of arsenic by CaO at such high temperature is rarely investigated. Understanding the reaction mechanism is conducive to control the arsenic emissions from coal-fired flue gas in furnace and promote arsenic capture at high temperature by adopting corresponding measures.

Considering that the decomposition temperature of possible product (calcium arsenate) derived from reaction between arsenic and CaO was only approximately 1400 °C. Therefore, in this work, the reaction temperature was controlled between 1000 °C and 1300 °C. SO_2 and NO, common components present in coal-fired flue gas, might react with CaO or affect the adsorption behavior of arsenic on CaO surface. Thus, SO_2 and NO were both investigated. Furthermore, the retention of arsenic included adsorption and oxidation process, and the latter played a more crucial role in arsenic capture. However, the mechanism regarding oxidation process has not been absolutely clarified and needed to be further illuminated. In this study, the corresponding experiments were designed and conducted, the reasonable result was obtained.

2. Experimental procedures

2.1. Materials

The chemical reagents used in the arsenic capture experiment and subsequent digestion process were hydrochloric acid, sodium hydroxide, arsenic standard liquid, calcium oxide, potassium borohydride and thiourea. All chemicals were analytical grade and could be used without further purification.

2.2. Adsorption experiments

Arsenic adsorption experiments were performed and the schematic diagram of experimental system was shown in Fig. 1. This system mainly included two portions: gaseous arsenic generation and adsorption (The system was constructed according to the literature [14]). In the first portion, the gaseous AsH_3(g) was provided by the liquid phase reaction among $NaAsO_2$, HCl and KBH_4 solution in a hydride generator (HG-01, Beijing east-west analytical instrument co. LTD). It was then mixed with carrier gas including N_2 (83 v/v%) as well as O_2 (17 v/v%)) and oxidized into As_2O_3(g) at 700 °C in the upper region of resistance furnace with two temperature zones. The required As_2O_3(g) concentration could be adjusted by peristaltic pump in the hydride generator. In the second portion, As_2O_3(g) derived from the first portion was carried with carrier gas and adsorbed by CaO located in the lower region of resistance furnace in the temperature range of 1000 °C to 1300 °C. The gaseous arsenic obtained by the method was more stable and easily controlled as compared to the conventional way (Gaseous arsenic was obtained by heating solid arsenic (As_2O_3(s)) directly).

In the experiment, 0.5 g CaO was weighed and put in a mullite boat. The mullite boat was attached to the lower part of the heated quartz tube. Hydride generator began to run when the temperature reached the specified temperature, the constantly arsenic vapor was produced with the assistance of O_2 as mentioned above. The adsorption process in the second process was then carried out in mullite boat. To investigate the effect of SO_2 and NO on arsenic capture, a certain amount of SO_2 (2000 ppm)/NO (500 ppm) was introduced into the carrier gas. The products were disposed after capture by acid digestion and the amount of arsenic captured by CaO was detected. Each experiment was repeated three times and the data shown in figures was average.

2.3. Analytical methods

The capture performance of CaO was evaluated by the amount of arsenic adsorbed which was detected by atomic absorption spectroscopy (AAS, AA-7020, Beijing east-west analytical instrument co. LTD). After wet digestion, the concentration of arsenic was tested by AAS and the capture capacity of CaO was also calculated as below:

$$A = \frac{C_{As} \times V_{As}}{M_{CaO}} \tag{1}$$

where A is the adsorption amount of CaO, mg/g; C_{As} denotes the

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