



Findings of proper temperatures for arsenic capture by CaO in the simulated flue gas with and without SO₂



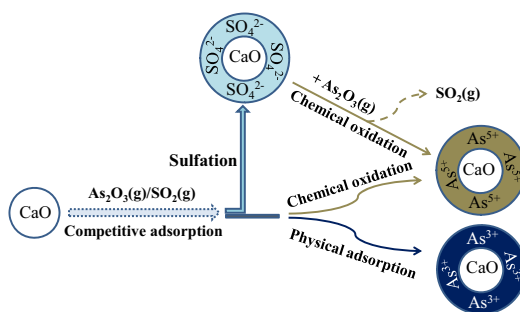
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HIGHLIGHTS

- Temperature increasing plays a positive role in arsenic capture below 1173 K.
- Arsenic capture is suppressed by CaO sintering at 1173 K and higher temperatures.
- SO₂ competes with arsenic vapors to react with CaO, inhibiting arsenic capture.
- The formed CaSO₄ is able to absorb arsenic vapors at high temperatures.
- As capture by CaO should be properly conducted around 873 K at the presence of SO₂.

GRAPHICAL ABSTRACT



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ABSTRACT

Arsenic emission during fuel combustion is of great concern due to its high toxicity. CaO shows a great capacity for arsenic capture and the addition of CaO is a promising way to suppress arsenic emission during fuel combustion. SO₂ in the flue gas could also react with CaO and might influence arsenic capture. It remains unknown which temperature is proper for CaO injection for arsenic capture considering the effect of SO₂. And the reaction mechanism is still unclear at different temperature range. In the present study, the capture of arsenic by CaO was investigated in simulated flue gas with and without SO₂ at temperatures ranging from 573 to 1323 K. The results show that arsenic capture was predominantly through physical adsorption at 573 and 723 K while was mainly carried out through chemical oxidation at higher temperatures. With temperature increasing, arsenic capture was enhanced. Nevertheless, the sintering of CaO particles occurred at 1173 and 1323 K. It is sure that arsenic capture was remarkably suppressed at these temperatures owing to the particles sintering. On the other hand, SO₂ competed with arsenic vapors to react with CaO. The enhancement of sulfate reaction strongly inhibited arsenic capture with temperature increased from 573 to 1023 K. At higher temperatures, the formed CaSO₄ was confirmed to be able to absorb arsenic vapors which could partly facilitate arsenic capture. Considering the sintering of CaO particles and the effect of SO₂, the proper temperature for arsenic capture should be around 873 K.

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1. Introduction

Arsenic is of high toxicity and excessive levels of arsenic exposure could cause serious health problems to both humans and ani-

mals [1–3]. Apart from the arsenic emission from natural sources, a considerable amount of arsenic is released into atmosphere from fuel (such as coal and municipal solid waste) combustion in China [4,5]. To meet the increasingly stringent environmental requirements, it is essential to control the atmospheric emissions of arsenic from fuel combustion.

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Arsenic is easily volatilized and is predominantly present in As^{3+} form as vapors (such as $As_2O_3(g)$) during fuel combustion [6,7]. As in the vapor phase, arsenic tends to concentrate in fine particles [8] which are hard to be captured by electrostatic precipitator, baghouse filter and other air pollution control devices. At high temperatures, the interactions between some inorganic minerals and arsenic vapors promote arsenic capture by forming various arsenates (As^{5+}) [9,10]. Addition of sorbents is a promising way to suppress the release of the easily volatilized trace elements [11–13]. These volatiles might react with the added sorbents and then were transformed into larger and easier captured particles [14,15]. Calcium compounds were usually used as sorbent for the control of trace elements release [16,17]. Zhao et al. [18] found that through the addition of calcium based sorbent, arsenic was enriched in larger ash particles through the formation of calcium arsenates (As^{5+}). In our previous study, the capacity of calcium compounds for arsenic capture was further confirmed during municipal solid waste incineration [19]. Moreover, calcium in various forms shows different capacity for arsenic capture. Experiments conducted by Sterling and Helble [20] demonstrated that CaO was more effective for $As_2O_3(g)$ capture than di-calcium silicate ($2CaO \cdot SiO_2$) and mono-calcium silicate ($CaO \cdot SiO_2$).

Although the reactions between CaO and $As_2O_3(g)$ have been widely investigated, the mechanism was still unclear and several divergent opinions need to be clarified. According to former researchers' studies, it is hard to get a proper temperature for CaO sorbent injection to remove arsenic from the flue gas. Jadhav and Fan [21] claimed that arsenic capture by CaO was enhanced with temperature increasing from 573 to 873 K while less arsenic was captured at higher temperatures from 973 to 1173 K. However, Li et al. [22] supported that temperature increasing from 873 to 1273 K stimulated arsenic capture. The forms of arsenic in the reaction products were detected in these two studies. It was supposed that the formation of various calcium arsenates ($CaAs_2O_7$ vs. $Ca_3As_2O_8$) was the main reason for the opposite conclusions.

On the other hand, arsenic has a strong affinity to participate in sulfide minerals especially in coal [23,24]. In the combustion process, the released SO_2 in the flue gas could also react with CaO [25]. So far, no identical conclusion has been given regarding the effect of SO_2 on the arsenic capture by CaO. Seames and Wendt

[26] found that SO_2 suppressed $As_2O_3(g)$ capture by Ca and/or Fe compounds in a 17 kW self-sustaining laboratory combustor. However, Li et al. [22] argued that the rate constant of arsenate reaction was much higher than that of sulfate reaction. The presence of SO_2 hardly affected $As_2O_3(g)$ capture at 1073 K in a thermo gravimetric analyzer reactor. In these experiments, the conditions were quite different especially for the reaction temperature. Thereby, the conclusions from these results were incomparable and further study should be carried out to confirm these conclusions.

In view of the problems mentioned above, one of the main objectives in this study is to clarify the mechanism of the interaction between CaO and $As_2O_3(g)$. Unlike the arsenic source supplied by heating $As_2O_3(s)$ in previous researchers' studies, the arsenic source was constantly provided by using a hydride generator in this work. The sorption experiments were carried out in simulated flue gas at temperatures ranging from 573 to 1323 K. Apart from the total amount of the captured arsenic, the speciation of arsenic in the adsorption products was investigated to further understand the pathway for arsenic capture. In addition, arsenic capture in the carrier gas with SO_2 was observed to illuminate the effect of SO_2 on the $As_2O_3(g)$ capture by CaO.

2. Experimental procedures

2.1. Materials

In the present work, CaO particles were prepared from the synthetic CaO through the smashing and sieving processes. The particle size of the used CaO is from 200 to 300 μm . The $As_2O_3(g)$ source was obtained through the oxidation of $AsH_3(g)$, which was constantly produced by using a hydride generator (HG-B, Beijing Shuguangming Electronics Instrument Co., Ltd.). For the production of $AsH_3(g)$, $NaAsO_2$ was firstly dissolved in 1 (v/v%) HCl solution and the prepared solution was used as arsenic standard solution. Then the $NaAsO_2$ solution was mixed with HCl solution and KBH_4 solution to produce $AsH_3(g)$. To generate constant concentration of $AsH_3(g)$, the flow rates of these solution were precisely controlled by a peristaltic pump (as shown in Fig. 1). Pure argon was used as carrier gas to put $AsH_3(g)$ into the reactor.

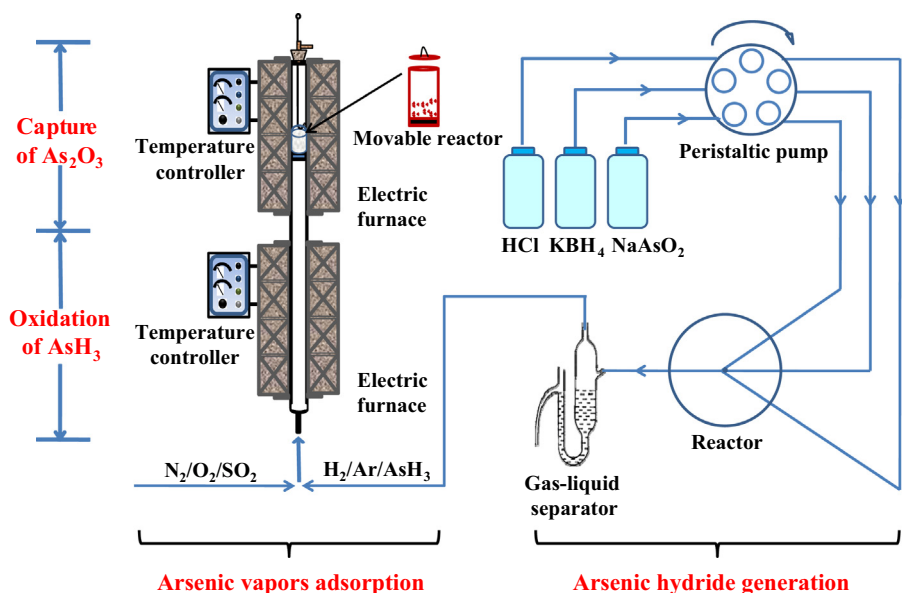


Fig. 1. The schematic of arsenic hydride generation and arsenic vapors adsorption set-ups.

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