



Full Length Article

Effect of ash composition on the partitioning of arsenic during fluidized bed combustion

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HIGHLIGHTS

- The transformation behavior of arsenic were affected by ash composition.
- The retention efficiency was affected by combustion temperature and addition ratio.
- Chemical adsorption may be the primary mechanism between arsenic and ash components.

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ABSTRACT

The effect of the ash composition on the redistribution of arsenic in a coal combustion system was determined via both experimental laboratory simulations and thermodynamic equilibrium calculations. The experimental study was performed in a fluidized bed reactor in combustion temperature range of 500–1000 °C. The results show that arsenic redistribution is strongly related to the chemical composition of coal in a combustion system. The presence of Fe, Ca, Mg, Al, Na and K favour arsenic retention, while Si causes an opposing effect. The arsenic retention on ash compositions is both temperature and amount dependent. With rising combustion temperature, the arsenic retention efficiency of Fe, Ca and Mg oxides augmented up to a maximum and then decreased, while those of Al, Na and K compounds continued expanding in the range of 500–1000 °C. Chemical adsorption may be the primary mechanism for Fe, Ca, Mg, K and Na, while the adsorption of arsenic by Al may be ascribed to both physical and chemical adsorption. Arsenic is captured in ashes as a result of the formation of stable arsenate compounds arising from interactions with the inorganic matrix. The negative arsenic retention effect by silica is attributed to reactions with ash compositions which promote arsenic capture and the reduction of the solid arsenic compounds.

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1. Introduction

Coal used for energy generation is identified as one of the largest anthropogenic contributors to environmental sensitively trace elements [1]. Toxic heavy metal elements such as As, Hg, Se, Pb and Cd are at least partially volatilized at high temperature and cannot be captured by atmospheric pollution control devices [2–5]. The total emissions of As, Hg, Se, Pb and Cd from coal combustion by energy generation in 2012 were estimated at approximately 406.4, 139.4, 538.6, 833.0 and 15.5 tons in China, respectively [6].

As a result of emerging environmental and health concerns, a series of environmental legislations and technologies are related with reducing these toxic elements in environment [7].

Arsenic and its compounds are known to be of greatly environmental and health concern due to its high toxicity and carcinogenicity [8,9]. Many studies have addressed that arsenic is associated with sulfide minerals, clay minerals, carbonate minerals and organic matter [10–16]. It has been reported that arsenic associated with sulfides and organic matter is volatilized during combustion, and subsequently released as vapor phase or particulate phase [2,17–19]. The elemental arsenic (As) and arsenic trioxide (As₂O₃) are supposed to the two probable forms of arsenic in the oxidizing flue gas environment [7,20]. For the chemical reactivity of elemental arsenic, As₂O₃ is regarded as the only species in the combustion environment [7]. Gas phase arsenic is preferred to

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deposit on fine particle which contains larger surface area and active cation sites in the cooling flue gas [2,19,21]. These arsenic-enriched fine particles are reduced limit by currently widely applied atmospheric pollution control devices (electrostatic precipitator and wet flue gas desulfurization), and partially emitted into environment. The arsenic removal efficiencies of electrostatic precipitator and wet flue gas desulfurization are estimated at about 86.2% and 80.4%, respectively [6]. Consequently, these emitted arsenic could be inhaled and deposited in lungs of humans and resulted in adverse health risks [8]. Meanwhile, many studies addressed that As_2O_3 in flue gas could react with active V_2O_5 sites and block the catalyst pores, and are prejudicial to selective catalytic reduction (SCR) installations [22]. Therefore, the study on gaseous arsenic control is of great environmental and industrial urgency.

The redistribution behavior of arsenic during combustion is dependent on many factors, including the association in coal, the interaction with other element both during and after combustion, the combustion conditions (temperature and boiler type) [18,23,24]. It has been reported that the partitioning behavior of arsenic is not only determined by the volatility of arsenic, but also affected by the interactions with different mineral components [25,26]. Thus, any improvement to the process that favour a more efficient retention of arsenic by mineral components is worthwhile. Recycling fly ashes or some of mineral components for the retention of mercury, arsenic and selenium has been addressed, and a positive relationship is found between fly ash calcium and iron concentrations and the corresponding arsenic retention concentration in the fly ash [27–30]. The ability of various mineral sorbents, including silica, alumina, kaolinite, limestone, bauxite for gaseous arsenic retention is well established in the literature, and illustrated that Ca-, Al and Fe-based sorbents are more efficient in arsenic retention during combustion process [20,27,31]. Although many previous studies have been conducted on gaseous arsenic retention by Ca, Al and Fe-based materials, the interactions between arsenic with other typical ash compositions (Si, Mg, Na and K) are extremely scarce [25]. Meanwhile, many previous studies determined the retention behaviors of arsenic by using pure arsenic compounds (As_2O_5 standard liquid and As_2O_3) [7,20,27]. These simulating experiments may not reflect the real retention behavior of arsenic during coal combustion due to the complex chemical reactions among fuel compositions [25]. Therefore, study on the interaction mechanism of arsenic with the mineral components of fly ash by co-combustion coal with ash compositions in a fluidized bed reactor may be useful for identifying the optimum technology for gaseous arsenic removal.

Thermodynamic equilibrium modelling could provide useful information for the interactions among two compounds, and have been widely applied to investigate the partitioning behavior of arsenic during combustion [31,32]. Nevertheless, the equilibrium modelling is calculated based on a particular condition (temperature, pressure and atmosphere), and are not considered about the complexity (association of arsenic in different type coal, size distribution of coal, mass transfer, interactions with other elements) in real combustion system [25]. Therefore, the thermodynamic equilibrium calculations were combined with experimentation to evaluate the possible reaction mechanisms between arsenic and different mineral components.

The main objectives of this study are to: (1) ascertain the effect of combustion temperature, ash composition type and amount on arsenic retention during combustion; (2) evaluate the arsenic retention mechanism by different ash compositions. The ultimate aim will try to identify the ash compositions and combustion conditions which could optimize the gas-phase arsenic control.

2. Experimental

2.1. Materials

The bituminous coal used for power generation in Pingwei Coal-fired Plant was selected and collected. The physico-chemical property of the selected coal including ash, proximate and ultimate analysis were determined by our previous study and summarized in Table 1 [3]. The ash yield of the selected bituminous coal was approximately 13.1 wt.% and labeled as medium-to-low ash. The concentration of arsenic was 3.24 mg/kg, which was similar to the weighted average value of Chinese coal (3.79 mg/kg) [33]. The selected coal was air-dried, grinded and sieved into 100–150 μm for subsequent analysis.

2.2. Combustion procedures

The laboratory-scale fluidized bed furnace with a height of 1500 mm and an inside diameter of 50 mm was used to investigate the retention behavior of arsenic during combustion with various ash compositions. The details of the experimental equipment and procedure were found elsewhere [3,24]. The furnace was made by stainless steel and heated by a batch of electrical resistances packed with ceramic fibers to thermal insulation. The fuel feeding rate, the excess air ratio (relative to the theoretical air), the fluidizing air flow, the secondary air flow and the fuel/air ratio were 0.2 kg/h, 40%, 1 Nm^3/h , 0.5 Nm^3/h and 0.714, respectively. The details of the combustion conditions were presented in Table 2. The admixture of coal and additives was pneumatically placed in a screw feeder, equipped with an agitation chamber and an insulated injector to ensure the uniform fuel feeding rate. The addition ratios of ash compositions were 1 wt.%, 3 wt.% and 5 wt.% of the selected coal. For the chemical instability of alkali metal oxides (K_2O and Na_2O) in environment, the oxides were substituted by the alkali metal nitrates ($NaNO_3$ and KNO_3). The weight percentages of the added ash compositions were calculated as oxides content. Six combustion temperature points including 500 °C, 600 °C, 700 °C, 800 °C, 900 °C and 1000 °C were conducted. The surface area, pore volume, pore diameter and particle size of the added ash chemicals were measured by an adsorption volumetric analyzer ASAP 2020 and Nano S laser particle size analyzer, respectively. The structure characteristics of the selected additives could be found in Table 3.

The feedstock was fed into the reactor when the temperature reached the desired level and attained at a steady state. The combustion procedure was maintained approximately 2 h. The bottom ash and fly ash were collected at each site simultaneously. In addition, in order to investigate the distribution behavior of arsenic in different particle size during different addition ratios, the fly ash were sieved by the standard Tyler sieve into three parts of particle size distribution (<38 μm , 38–200 μm , and >200 μm). In order to ensure the precision and accuracy of the experimental results, the sampling for each experimental condition were repeated in triplicate. Once the relative deviations of the data from these samples were more than 20%, another samplings were required.

The retention efficiency (RE) could illustrate the capture behavior of additives during combustion effectively. Generally, arsenic may be captured by ash compositions via both physisorption and chemical reactions. The retention efficiency was calculated as described below:

$$RE (100\%) = \frac{V_c - V_{ad}}{V_c} \times 100 \quad (1)$$

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