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## Retention mechanisms of ash compositions on toxic elements (Sb, Se and Pb) during fluidized bed combustion



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#### ABSTRACT

Both laboratory simulation experiments and thermodynamic equilibrium calculations were conducted to illustrate the retention mechanisms of ash components on three toxic elements (Sb, Se and Pb). The experiment was carried out in a fluidized bed reactor with the combustion temperature in the range of 500-1000 °C. The results shown that the redistribution behavior of these toxic elements was greatly proportional to the combustion temperature, ash components and amounts. The adding of Fe, Ca, Mg, Na and K favour the retention of Sb and Se, while the presence of Si was represented against Sb and Se capture. Si, Al, Fe, Na and K have an appreciable effect on the retention of Pb, while the Pb capture by Ca and Mg was limited. The partial retention of these elements in ashes may be attributed to both physisorption and chemisorption. Among them, the formation of non-volatilized compounds may be the primary contribution for the retention of Sb, Se and Pb (except for Al). The negative Sb and Se captured by Si may be ascribed to the interactions between Si and other components which promote the retention of Sb and Se. The highly Pb retention by Si and Al in ashes may be explained by the formation of PbO·SiO<sub>2</sub>, PbO·Al<sub>2</sub>O<sub>3</sub> and PbO·Al<sub>2</sub>O<sub>3</sub>·2SiO<sub>2</sub>. Fe- and Al-based materials may be supposed to the appreciate additives for the capture potential of all the selected elements.

#### 1. Introduction

Coal is known as the most significant source of energy generation for its affordability and reliability. Nevertheless, coal combustion faces many serious technical limitations in terms of the environmental considerations [1]. One of the primary concerns of coal combustion is the substantial amount of toxic elements, which can partially or totally volatilized at high combustion temperature and escaped from currently atmospheric pollution control devices [2–6]. The total emissions of Hg, As, Se, Pb, Cd, Cr, Ni, and Sb in 2010 were approximately 118.54, 335.45, 459.4, 705.45, 13.34, 505.03, 446.42, and 82.33 tons (t) in China, respectively [7]. The emission of these toxic elements continued its remorseless rise undoubtedly due to the lack of effective control devices and the mounting consumption and requirement of coal [8–11]. It is unquestionable that the large emissions of these toxic elements could cause a serious impact on eco-environment and mental health [12]

Antimony, selenium, lend and their compounds are regarded as environmentally sensitive toxic elements for their toxicity and bioaccumulation [13]. Several strategies have been developed to investigate the associations of these elements in coal, and suggested that these elements were mainly existed in sulfide minerals, clay minerals and organic matter [14–16]. As combustion proceeds, toxic elements which were trapped in an organic matrix or sulfide minerals may be released and transferred to the gas phase, finally display enrichment in the fine particle via vaporization-condensation pathway or emit as vapor phase directly [3,17]. The oxides (SbO, SeO<sub>2</sub>, and PbO) were regarded as the primary forms of these elements in the oxidizing environment [18–20]. These elements in gaseous and fine particulates were difficult to captured by currently atmospheric pollution control devices (electrostatic precipitator, fabric filters, wet flue gas desulfurization), and finally partially entered into environment. Approximately 95% of total installed capacity coal-fired units were equipped with electrostatic precipitator, while 5% were used fabric filters or combined electrostatic precipitator and fabric filters in China up to 2010 [7]. The Sb, Se and Pb capture efficiencies were 83.46, 73.78 and 97.17% for electrostatic precipitator, 94.25, 65.00 and 99.00% for fabric filters, and 82.10, 74.87, 78.40% for wet flue gas desulfurization, respectively [7]. Subsequently, these uncontrolled elements have considerably longer atmospheric lifetimes and could be inhaled, and finally lead to a serious

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impact of mental health [21–23]. Meanwhile, these toxic elements were primary enriched on the surface of the fine particles, increasing the potential reactivity and biological impact [24,25]. Therefore, it is of greatly urgent for the capture of these volatized elements during coal combustion.

The methods of controlling toxic elements during coal combustion were classified into three categories: pre-control (coal preparation, coal washing, and coal waste slurry technology), mid-control (fluidized bed, the use of additives and bag filter technology), and post-control (dust collectors, flue gas capture device, and multi-stage purification device) [25,26]. Much effort has been expending in studying the redistribution behavior of toxic element during coal combustion, and suggested that the major-factors effecting toxic element emission from coal-fired plants were not only the volatility and associations of the element during combustion, but also the interactions between element and ash compositions [27-30]. Since the organically associated elements and the host minerals trapped in an organic matrix cannot be removed from coal by conventional coal cleaning techniques, the pre-control methods exhibit a limited contribution for toxic element retention. The midcontrol techniques, especially the use of additives, were regarded as the most effective pathway for toxic element capture and have received fairly intensive study [31-36]. Many mineral additives, including silica, alumina, kaolinite, limestone, bauxite, and modified fly ashes were used for retention of mercury, arsenic, selenium, cadmium and lead, and reported that there was a positive correlation between mineral components (Ca, Fe and Al) in feedstock and toxic element enriched in fly ash [32,33,35,37-40]. Therefore, adding ash composition may be effective for the capture of toxic element. Although many studies have addressed the toxic element retention efficiencies by different ash compositions, the reports on the retention mechanisms of these ash compositions (especially for Na, K and Mg) are extremely scarce [41]. In addition, these previous studies investigated the toxic element capture behavior via aqueous solutions of element (As, Sb, Se, Cd and Pb) oxides, investigations based on the coal and additive bleeding were extremely few [18,25,32]. Therefore, identification of the interaction mechanisms between ash compositions and toxic elements by using both simulated coal-additive combustion experiments in a fluidized bed reactor and thermodynamic equilibrium calculations may aid the development of thermodynamic models of toxic element during coal combustion, which are particularly useful for novel toxic element control techniques.

Fluidized bed combustion is regarded as an effectively clean coal technology for its benefit in fuel flexibility, combustion efficiency, boiler sintering, reduction of  $SO_2$  and  $NO_x$  [42]. Both quantity and capacity of the fluidized bed boilers are steadily increasing over the past decade, and this tendency is expected to continue for the severely environmental laws and high combustion efficiency. The combustion conditions, ash characteristics, and control devices are different between pulverized coal and fluidized bed combustion, and subsequently resulted in the difference in toxic element partition behavior [17]. There have been many investigations into the distribution, enrichment, transformation and environmental impacts of toxic elements during fluidized bed combustion [17,42,43]. Duan et al. (2016) investigate the partition of trace elements in a 2.5 MWth fluidized bed combustor, and reported that most of the elements were existed in bottom and fly ash, while small amounts of As, Cd, and Pb were entered into atmosphere along with fine particulates [42]. Jiao et al. (2013) analyzed the leaching characteristics of arsenic in fly ash from fluidized bed coal combustion, and found that mobility potential of arsenic in fly ash is largely determined by the pH of the leachate [43]. Nevertheless, the study on the retention mechanism of toxic elements by ash compositions was scared. The main objectives of this present work are to: (1) determine the association and volatility of the selected toxic elements (Sb, Se, and Pb) in coal, (2) investigate the effect of ash components, combustion temperatures, and additive amounts on the retention of the selected elements, (3) reveal the interaction mechanisms between the selected elements and ash compositions.

#### 2. Experimental

#### 2.1. Coal sample

The bituminous coal selected in this study was prepared from Pingwei Coal-fired Plant, which was used for power generation. The physico-chemical characteristics including proximate, ultimate, ash and mineralogical properties were described in detailed at the previous study [44]. Briefly, the selected coal was regarded as typical medium to low bituminous coal with an ash yield of 13.1 wt%. The volatile matter and fixed carbon were 29.0 and 56.1 wt%, respectively. SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> were the predominantly chemical oxides, which were 53.1 and 32.7 wt %, respectively. The contents of Fe<sub>2</sub>O<sub>3</sub> and CaO were 4.31 and 3.13 wt %, respectively. The mainly minerals in the selected coal were kaolinite, quartz, calcite, pyrite and illite, which were the commonly minerals in coal. The concentrations of Sb, Se and Pb were 3.36, 4.61 and 7.53 mg/kg, respectively. In comparison with the weighted average values of Chinese coal (0.84, 2.47 and 15.1 mg/kg for Sb, Se and Pb, respectively) reported by Dai et al. (2012) [45], the contents of Se and Sb were higher.

#### 2.2. Combustion procedures

The transformation behavior of toxic elements (Sb, Se and Pb) during combustion with ash compositions was conducted on a laboratory-scale fluidized bed furnace which described detailed in the previous study [44]. The laboratory-scale fluidized bed furnace was 1500 mm in height and with an inside diameter of 50 mm. The fuel feeding rate, the excess air ratio (relative to the theoretical air), the fluidizing air flow, the seconding air flow and the fuel/air ratio were described in the previous study [42], which were 0.2 kg/h, 40%, 1 Nm<sup>3</sup>/h, 0.5 Nm<sup>3</sup>/h, and 0.714, respectively. Various adding ratios of ash compositions including 1 wt%, 3 wt% and 5 wt% of the coal were applied. As mentioned in the previous study [42], the alkali metal nitrates (NaNO3 and KNO3) were used due to the chemical instability of alkali metals oxides (K2O and Na2O). The weight percentages of the added alkali metal nitrates were transformed as oxides content. The microstructure characteristics including surface area, pore volume and pore diameter of the selected additives were also described in the previous study [44]. Briefly, the surface areas of SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and MgO (45.58, 85.32 and 68.76  $m^2/g$ , respectively) were higher greatly than that of Fe<sub>2</sub>O<sub>3</sub> and CaO (14.27 and 7.44  $m^2/g$ , respectively), the pore diameters of SiO<sub>2</sub>, Fe<sub>2</sub>O<sub>3</sub> and CaO (14.40, 23.65, 13.68 nm, respectively) were higher than that of Al<sub>2</sub>O<sub>3</sub> and MgO (7.24 and 8.27 nm, respectively). The various microstructure characterizations of the additives may influence their retention behavior on toxic element.

The combustion was carried out at the temperature of 500–1000 °C. The feedstock was entered into the reactor when the temperature reached the designed requirement. The whole combustion procedure was conducted approximately 2 h. The combustion residues including bottom ash and fly ash were obtained separately. Meanwhile, the fly ash was sieved into three different size distributions (< 38  $\mu$ m, 38–200  $\mu$ m, and > 200  $\mu$ m) to determine the redistribution behavior of toxic element during combustion with different additives. Each combustion condition was conducted in triplicate to ensure the precision and accuracy.

The retention of toxic element could be occupied by both physisorption and chemical absorption. The retention efficiency (RE) was defined as the capture behavior of additives which could be calculated as the following equation:

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

where V<sub>c</sub> and V<sub>ad</sub> were the volatilization ratios of toxic element during

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