



Full Length Article

Enrichment characteristics, thermal stability and volatility of hazardous trace elements in fly ash from a coal-fired power plant



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ABSTRACT

Release of hazardous trace elements (HTEs) during re-use of coal-fired fly ash involving heat treatment process can cause health risks for on-site workers and contaminate the surrounding environment. Enrichment characteristics, thermal stability and volatility of five HTEs (Hg, As, Pb, Cr, Mn) in fly ash collected from a 660 MW coal-fired power plant were investigated. Brunauer-Emmett-Teller (BET), scanning electron microscope-energy dispersive spectrometer (SEM-EDS), and X-ray power diffraction spectrometry (XRD) were used to characterize pore structure and chemical composition of the coal-fired fly ash (CFA) samples. Results show that the coal belongs to bituminous, containing low content of S, Cl, and the HTEs analyzed. CFA samples have low content of Hg (0.04–0.20 mg/kg) and As (3.0–8.7 mg/kg), and high content of Mn (782.46–1115.85 mg/kg). Burnout degree and specific surface area result in the decreasing HTEs concentration in CFA samples when increasing the particle size. Relative enrichment index (REI) of HTEs in the raw CFA are in the order of Mn > Cr > Pb > 1 > As > Hg. Increase in particle sizes leads to decrease of the REI value, implies that HTEs are enriched more in finer fractions. Thermal stability of HTEs in CFA samples becomes weaker at higher temperature. The smaller the particle size is, the worse the thermal stability becomes. Volatilization fractions of Hg, As, Pb, Cr, and Mn in CFA samples at four temperatures (350 °C, 550 °C, 750 °C, 950 °C) range 88.35%–99.50%, 2.32%–23.17%, 4.08%–22.59%, 20.36%–34.43%, and 1.21%–23.43%, respectively. The derivative of the concentration-fitted curve to the temperature in the range of 20–900 °C at heating rate of 10 °C/min indicates volatility of HTEs in CFA follows the decreasing order of Hg > As, Cr > Mn, Pb.

1. Introduction

Hazardous trace elements (HTEs), namely Hg, As, Cr, Mn, Pb, Ba, etc., emitted from coal combustion can cause harm to the environment by contaminating air, water, soil and can cause human health hazards [1–3]. U. S. Clean Air Act Amendments (CAAA) listed 11 elements including Hg, As, Cr, Mn, Ni, Se, Be, Cd and Co as the key toxic air pollutants in 1990; in which Hg, As, Cr, Pb, Se and Cd were chosen as priority elements [4,5]. Some HTEs, like As, Pb, Hg, Ni, and Cd, have also been considered as prime environmental concerns by the European Union and the Canadian Environmental Protection Agency. In China, the State Council has approved the 12th five-year plan for comprehensive prevention and control of some HTEs pollution in 2011 [6]. With huge amount of coal consumption, coal-fired power plants for electricity generation are considered as one of the major anthropogenic emission sources of HTEs [7,8]. During coal combustion, some HTEs

existed in coal volatilize and react with surrounding environment including flue gas components and fly ash. With flue gas cooling, HTEs undergo homogeneous nucleation, condensation reaction, or adsorption. Finally, HTEs are emitted in gaseous and particulate forms in the flue gas, and part of them occur in bottom ash [9]. Field tests on coal-fired power plants have shown that most HTEs, except Hg, are mainly distributed in fly ash removed from particulate collection devices such as electrostatic precipitator (ESP) or fabric filter (FF) [10–13]. This indicates that the particular attention should be given to the HTEs pollution during treatment of the coal-fired fly ash.

The annual generation capacity of coal-fired fly ash in China, the USA and India in 2015 was about 580, 130, 190 million tonnes, respectively [14]. Based on the physical, chemical and mineralogical properties, coal-fired fly ash can be used for different purposes such as applied in soil amelioration, construction industry, ceramic industry, catalysis, environmental protection, depth separation, zeolite synthesis,

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and valuable metal recovery [15]. According to the annual comprehensive resource utilization report (2012) released by the National Development and Reform Commission (NDRC) of China, the generation and utilization amount of coal-fired fly ash was 540 and 367 million tonnes in 2011, respectively. In all the sections of utilization, the top three are cement (41%), brick and tiles (26%) and concrete (19%). However, when the coal-fired fly ash is used as a raw material in a production process like cement, brick and tiles, the high temperature treatment process is unavoidable [16,17]. It can lead to release of HTEs previously existed in coal-fired fly ash, which can pose toxic occupational health risks to on-site workers and neighbors. Huang et al. [18] investigated the effects of some vital flue gas components and mineral sorbents on the partitioning behavior of four major HTEs (Cd, Pb, Zn and Cu) which are often present in municipal solid waste (MSW), finding that presence of HCl promotes evaporation of the four HTEs while SiO_2 - or Al_2O_3 -containing minerals may benefit for their control. Wu et al. [19] prevented the evaporation of HTEs (Pb, Zn and Cd) by controlling their chemical speciation in MSW incineration fly ash, results of which indicated phosphate chemical pretreatment resulted in the least evaporation of HTEs in the fly ash. Hu et al. [20] compared the CaO's effect on the fate of HTEs (Pb, Cr, Zr, Cu, Zn and Cd) during thermal treatment of two typical MSW incineration ashes in China. It showed that conversion of Ca to aluminosilicates occurred especially at 1323 K, which promoted the HTEs immobilization and decreased their volatile fractions. In addition, Hu et al. [21] found that part of arsenic was stabilized in the ash matrix including some un-oxidized As(III) which remained stable even when the MSW incineration ash was heated at 1323 K. Nowak et al. [22] investigated the influence of chloride addition (zero to 200 g Cl/kg as added as CaCl_2 , MgCl_2 , or NaCl) on the trace elements (Pb, Cr, Cd, Zn, Ni and Cu) removal from MSW fly ash by treating their mixture in a muffle oven (800–1200 °C, 20 h) and a laboratory-scale rotary reactor (1000 °C, 60 min), results of which showed that Cd, Pb, and about 10% Cr were either present in ash in volatile form or also reacted with ash inherent Cl, while CaCl_2 and MgCl_2 were more effective for the evaporation of the most heavy metals than NaCl. All these above mentioned studies focused on release behavior of some trace elements with relatively low volatility such as Pb, Cr, As in the MSW incineration ash; in which the mineral compositions in MSW incineration fly ash and coal-fired fly ash are different, especially the Cl content. However, the studies related to the release of HTEs in coal-fired fly ash are very little, thus, there is a need to investigate on the release behavior of HTEs from coal-fired fly ash.

HTEs can be classified into three groups according to the partitioning behavior, which has been recognized by many researchers [23,24]. Group 1: have high volatility and are mainly emitted in gaseous state, such as Hg, Br, Cl, etc. Group 2: volatilize and be prone to adsorb or deposit on the surface of small particles with the decrease in flue gas temperature, such as As, Pb, Cd, Cu, and Zn, etc. Group 3: do not vaporize and be equally distributed in bottom ash and fly ash, such as Cr, Mn and Co, etc. Thus, HTEs, namely Hg, As, Pb, Cr and Mn, representing these three groups were chosen in this study. Enrichment characteristics, thermal stability and volatility of HTEs in fly ash with three different particle sizes were investigated. The main objective is to understand the release behavior of HTEs in coal-fired fly ash, which can provide guidance for the effective control of HTEs during re-use of coal-fired fly ash involving heat treatment process.

2. Experimental

2.1. Coal-fired fly ash (CFA) samples

The coal-fired fly ash (defined as CFA0) was collected from ESP (the first ESP in series) of a 660 MW coal-fired power plant. This plant was equipped with selective catalytic reduction (SCR), ESP, wet flue gas desulfurization (WFGD), and wet electrostatic precipitator (WESP) along the flue gas path. The coal used belongs to the bituminous. The

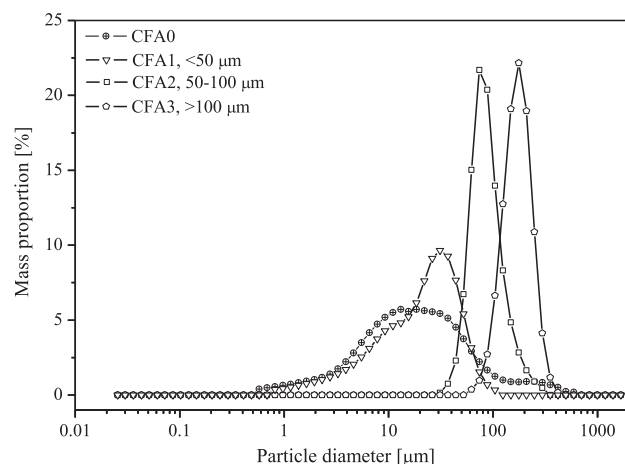


Fig. 1. Size distribution of the CFA0, CFA1, CFA2, and CFA3.

Laser particle size analyzer (Microtrac S3500, US) was used to obtain the particle size distribution of CFA0, result of which is shown in Fig. 1. Based on the size distribution, three kinds of particle sizes, namely < 50 μm (defined as CFA1), 50–100 μm (defined as CFA2), > 100 μm (defined as CFA3) were acquired by the mechanical screening method. The size distribution of CFA1, CFA2, and CFA3 is also shown in Fig. 1.

2.2. Determination of Hg, As, Pb, Cr and Mn in the CFA samples

The mercury analyzer named Milestone DMA80 was used to determine concentration of Hg in the CFA, detection limit of which was 0.2 ppb. For determining As, Pb, Cr and Mn in the CFA, they were firstly digested in a mixture of acids (HNO_3 , HCl, HF, HClO_4) and then measured by the inductively coupled plasma - mass spectrometry (ICP-MS, Agilent Technologies 7700x, US).

2.3. Thermal stability and volatility of HTEs in the CFA samples

To explore the thermal stability of HTEs in CFA with different particle diameters, CFA1, CFA2, and CFA3 were placed in a muffle furnace at 350, 550, 750, and 950 °C for 4 h, respectively. To study the volatility of HTEs in CFA samples, CFA1, CFA2, and CFA3 were placed in a tube furnace with a heating rate of 10 °C/min. Then, samples were taken out at the temperature of 150, 250, 350, 450, 550, 650, 750, 850, 950 °C, respectively. The concentration of Hg, As, Pb, Cr and Mn in the three CFA samples under each situation was determined.

2.4. Relevant characteristics of the CFA samples

To determine the unburned carbon (UBC) content in the CFA, the samples were dried at 102 °C for 8 h to remove moisture. The dried samples were then placed in a muffle furnace at 850 °C for 3 h. The ratio of the weight difference between the dried and burned samples to that of dried samples was defined as the UBC content [25]. In this study, the UBC contents of CFA0, CFA1, CFA2, and CFA3 are 11.76%, 15.59%, 11.43%, 9.65%, respectively, indicating the quality of CFA samples is in the order of CFA3 > CFA2 > CFA0 > CFA1 based on the burnout degree.

The Brunauer-Emmett-Teller (BET) surface area of CFA samples was determined using an automatic specific surface area and pore analyzer (ASAP-2020M, Micromeritics, US). Combination of scanning electron microscope (SEM, Hitachi s-4800 microscope, Japan) and energy dispersive spectrometer (EDS, Thermo Noran SYSTEM7, US) was used to characterize pore structure and chemical element of the surface. X-ray powder diffraction spectrometry (XRD, Shimadzu Corporation XD-3A, Japan) was used to detect the mineral composition. The XRD patterns

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