



Partitioning and transformation behavior of toxic elements during circulated fluidized bed combustion of coal gangue



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HIGHLIGHTS

- The relative distributions of toxic elements in the coal gangue power plant are studied.
- The toxic elements could be classified into two categories according to their volatilization.
- The transformation behaviors of toxic elements are analyzed systemically.

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ABSTRACT

A study on toxic elements behaviors from a 330 MW coal gangue circulated fluidized bed co-combustion power plant equipped with electrostatic precipitators (ESPs) was performed. Simultaneous sampling of feed coal, bottom ash, fly ash and flue gas were implemented. Sequential chemical extraction was taken to investigate the transformation behaviors of toxic elements. The relative distribution and partitioning behavior of toxic elements in the power plants were analyzed systemically. The toxic elements can be divided into two groups. Cluster one, whereby high volatile tendencies are represented by As, Cd, Cu, Pb, Se and Sn which have a volatilization ratio more than 20% and are mainly enriched in fly ash. These elements are primarily associated with sulfide minerals. Cluster two, represented by Co, Cr, Mn and V, have low volatilization rate ($\leq 5\%$) and are equally distributed between bottom ash and fly ash. In addition, Bi, Ni and Zn may be located between Cluster one and Cluster two. The variation of modes of occurrence of toxic elements could lead to the difference of transformation behaviors during the combustion of feed coal. Most toxic elements transformed and partitioned into other fractions from Fe–Mn oxides during combustion.

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1. Introduction

Coal gangue is the major exploiting residues (approximately 10–15% of coal production) produced during coal mining, which contains high ash content, low carbonaceous composition and low calorific value [1]. With the excellent heat and mass transfer properties, circulated fluidized bed combustion technology developed for co-firing coal gangue with coal have been steadily increasing in both quantity and capacity over the past decade [2,3]. Generally, the mixed fuel contains coal gangue and coal with a

blending ratio of 2–3:1. According to incomplete statistics collected up to 2010, there are more than 120 coal gangue circulated fluidized bed co-combustion power plants in China. Nevertheless, there are a large amount of toxic elements (As, Be, Cd, Co, Cr, Cu, Mn, Ni, Se, Pb, Sn, V, Zn, etc.) in feed coal. During combustion, with excellent heat and complex physic-chemical reaction, toxic elements could release from feed coal and enter atmosphere [4–10]. In view of the great demand in coal gangue use, an understanding of the behavior of chemical species in coal gangue during combustion and environmental transportation is of critical importance.

Extensive research on the toxic elements transformation behaviors in pulverized coal combustion system reported that the behaviors of toxic elements are determined by the coal composition, combustion atmosphere, physic-chemical characteristic and combustion technology [11–16]. Vassilev and Vassileva reported that

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toxic elements in coal could exist in both organic and inorganic constituents [5]. Querol et al. demonstrated that the trace element behaviors are mainly dependent on the coal particles, ash content and mineral composition [15]. There are few studies on toxic elements behavior in circulated fluidized bed co-combustion systems burning high ash content fuels and investigations are predominantly on relative distribution and partitioning of toxic element [17–19].

In addition, it is well known that the ash yield of coal gangue is higher than coal. Many studies have reported that toxic elements had a positive correlation with ash yield, indicating an inorganic affinity [20–23]. It is thus of great interest to analyze the mineral characterizations involved in coal gangue co-combustion. Generally, the determination of total toxic element content in feed coal and combustion residues could provide important information on the volatilization of toxic elements [24–27]. However, the measurement of total toxic element and volatilization behavior may not be able to provide the information about the transformation behaviors of toxic element associations effectively [22,24]. Therefore, it is important to implement chemical partitioning research on toxic elements in feed coal and combustion residues to assess the transformation characteristic accurately.

The sequential extraction method was organically employed to assess the mode of occurrence of toxic elements in soil samples and was later modified to study coal [28]. Based on sequential extraction procedures, the mode of occurrence of toxic element can be classified into five fraction: exchangeable, carbon bound, Fe–Mn oxides bound, organic matter bound and residual. Therefore, combination with correlation coefficient, the sequential extraction method was preformed to determine the transformation characterization of toxic elements' association in the coal gangue/coal co-fired power plant.

The main objectives of the present study are to determine (a) the transformation behavior of mineral composition during coal gangue circulated fluidized bed co-combustion, (b) the relative distribution, partitioning and transformation behavior of toxic elements in the coal gangue/coal co-fired power plant.

2. Materials and methods

2.1. Plant description

The study was carried out on a full-scale circulated fluidized boiler in Huainan, China. The unit is a novel power plant, which is fueled with coal gangue, coal and coal slime in a mixture ratio of 2:1:1. The plant has two independent coal power generation units, each of which is installed with 330 MW capacity. The boiler conditions are kept as stable as possible during the combustion, the bed temperature is approximately 900 °C. The coal used for combustion is mined from the Huainan Coalfield, which is classified as bituminous coal. The coal gangue is the by-product during mining activity, and the coal slime is the rejected materials during coal washing. Proximate and ultimate analysis of the combustion feed coals are presented in Table 1.

2.2. Sample collection

The samples were collected in one of two 330 MW coal-fired plants (Fig. 1). The flue gas samples were sampled isokinetically from the stack sampling point (at the top of stack) with a sampling duration of 1 h, following the American Standard Method EPA 29 [29]. The toxic elements in the gas phase were trapped in the aqueous acidic solution (10% H₂O₂ and 6% HNO₃) kept in impingers. Flue gas sampling conditions are shown in Table 2. The recovered samples were analyzed for As, Be, Cd, Co, Cr, Cu, Mn, Ni, Pb, Se, Sn, V and Zn by inductively coupled plasma mass spectrometry (ICP-MS).

For solid material sampling, 250, 40 and 60 kg of feed coal, bottom and fly ash were collected at each site for each sample simultaneously in step with the sampling of flue gas (1 h). To obtain representative samples, the samples were quartered and crushed prior to pass through a 150-mesh sieve in order to homogenize for the subsequent analysis.

2.3. Mineral analysis

The mineralogical characteristics of the feed coal, bottom and fly ash were analyzed using a PHILIPS X'Pert PRO X-ray powder diffraction (XRD) with Cu K-alpha radiation, and the patterns are recorded over a 2θ interval of 10–65°, with a step increment of 0.01°.

2.4. Chemical analysis

The powdered solid samples (feed coal, bottom ash, fly ash) and standard reference materials were digested using an acid mixture (HCl:HNO₃:HF = 3:3:2) in a microwave oven in Teflon beaker at 120 °C maintained for 20 min followed by 180 °C for 25 min. After digestion, HBO₃ was added to neutralize the remanent HF. Finally, the digestion solution was filtered through paper membrane and quantified to 25 ml with ultrapure water for the subsequent analysis. This sample extraction procedure has been successfully used in several previous studies [19,30–32]. The content of toxic elements (As, Be, Cd, Co, Cr, Cu, Mn, Ni, Pb, Se, Sn, V and Zn) in feed coal, bottom ash, fly ash samples were measured by ICP-MS.

The accuracy of analytical method was calibrated by standard reference materials NIST 1632b (coal) and GBW07406 (GSS-6) (soil), the recovery (92.6–102%) for all the selected toxic elements in standard reference materials were within the range of the certified values. The recovery is 93.2–105% when solutions with known toxic elements content are used. Thus, it could deduce that there was no toxic elements loss during digestion procedure. Meanwhile, background contamination was monitored by reagent blanks, which was digested simultaneously with the samples. The acceptable precision was within ±5 wt.% for toxic elements. If the disparity between the calibrated values and the standard solution was more than 5%, the apparatus needed to recalibrate. In order to guarantee the precision and accuracy of the experimental data, all the measurements for each sample were repeated three times. The relative standard deviations of the three times analysis were ranged from –1.53% to 2.47%, below the control level of ±5 wt.%.

Table 1
Proximate and ultimate analysis of the feed coal.

Proximate analysis, dry basis (wt.%)	Moisture	Ash yield	Volatile matter	Calorific value (MJ/kg)	
Feed coal	1.78 ± 0.09	48.5 ± 4.2	50.3 ± 5.6	11.8 ± 1.6	
Ultimate analysis, daf (wt.%)	C	H	N	O	S
Feed coal	30.2 ± 1.3	1.96 ± 0.07	0.62 ± 0.08	6.33 ± 0.32	0.46 ± 0.04

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