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Alteration behavior of mineral structure and hazardous elements during combustion of coal from a power plant at Huainan, Anhui, China[☆]

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ABSTRACT

The alteration behavior of minerals and hazardous elements during simulated combustion (100–1200 °C) of a raw coal collected from a power plant were studied. Thermogravimetric analysis indicated that there were mainly four alteration stages during coal combustion. The transformation behavior of mineral phases of raw coal, which were detected by X-ray polycrystalline diffraction (XRD) technique, mainly relied on the combustion temperature. A series of changes were derived from the intensities of mineral (e.g. clays) diffraction peaks when temperature surpassed 600 °C. Mineral phases tended to be simple and collapsed to amorphous glass when temperature reached up to 1200 °C. The characteristics of functional groups for raw coal and high-temperature (1200 °C) ash studied by Fourier transform infrared spectroscopy (FTIR) were in accordance with the result obtained from XRD analysis. The volatilization ratios of Co, Cr, Ni and V increased consistently with the increase of combustion temperature, suggesting these elements were gradually released from the organic matter and inorganic minerals of coal.

1. Introduction

Coal is the primary fuel resource for global electricity production. International Energy Agency (IEA) predicts that coal will continue to be the predominant fuel for electricity generation over the next two decades (IEA, 2017). According to the IEA report, thermal coal production growth rate likely ranges between 0.08% per year and 1.75% per year over 2012–2035 (56–132 million tonnes per year) (IEA, 2017). Domestically, China relies on coal to meet ~70% of its total energy need, and coal production is expected to increase in the near future (IEA, 2017; Bai et al., 2018; BP, 2018; Dai and Finkelman, 2018). At present, coal-fired power plants produced the majority of electricity used in China (Mishra et al., 2016). During coal combustion, minerals would undergo thermal decomposition, fusion, disintegration and agglomeration (Civeira et al., 2016a, b, c; Sehn et al., 2016; Oliveira et al., 2017, 2018). Hazardous elements in minerals and organic fractions of coal are liberated and distributed into combustion products, and then released into the environment (Swaine, 1990, 2000). Understanding the alteration behaviors of minerals and hazardous elements during coal combustion are crucial to predict their environmental fates (Rajabzadeh et al., 2016; Rodriguez-iruretagoiena et al., 2016; Saikia et al., 2016; Schneider et al., 2016; Agudelo-Castañeda et al., 2016, 2017; de Vallejuelo et al., 2017; Dutta et al., 2017; Gredilla et al., 2017).

Twenty-six (As, Cr, Cd, Hg, Pb, Se, B, Mn, Ni, Cu, V, Zn, Co, Sn and others) elements in coal are considered to be of environmental and health concern (Swaine, 2000). Hazardous elements in Chinese coals have drawn much more attention because huge amounts of coals have been burned in coal-fired power plants. This has released large fluxes of hazardous elements into the surrounding environments and caused adverse effects on human health (Dai et al., 2007, 2012; Finkelman and Tian, 2018). The abundance and partitioning behavior of the elements in combustion products depend on the sources and types of coal, modes of occurrence and volatilization/condensation characteristics of the elements and the

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operating conditions of power plants (Vassilev et al., 2009; López-Antón et al., 2011; Singh et al., 2013).

Previous studies mainly focused on coal and combustion products collected at coal-fired power plants, and the behaviors of mineral alternation and elemental volatility during coal combustion are affected by multiple factors (Quispe et al., 2012; Tang et al., 2012; Oliveira et al., 2014; Silva et al., 2014). Here, we study how the temperature affect the mineral alternation and elemental volatility using a simulated coal combustion device by controlling the temperature. We analyzed the thermogravimetric behaviors, typical mineral phases and functional groups of the combusted coal under different temperatures. The change of volatility of several hazardous elements (Co, Cr, Ni and V) with temperature was illustrated in detail.

2. Materials and methods

2.1. Samples

The experimental raw coal sample was a Permo-Carboniferous bituminous coal collected from a coal power plant, Huainan city, Anhui Province, China (Tang et al., 2013). Three replicates of raw coal sample was collected, which were air-dried and crushed to pass through 120-mesh sieve before analysis.

2.2. Combustion procedure

Twelve combustion temperature points were selected between 100 °C and 1200 °C with 100 °C interval. Approximate 5 g of raw coal sample was placed in a muffle roaster where the temperature was increased to the desired points following by 30 min dwelling. Then, the heated coal sample was taken out from the muffle roaster and cooled down to the room temperature in a dryer. We use the following notations to name the raw and heated coals: S-A (experimental raw coal), S-B (coal heated at 100 °C), S-C (coal heated at 200 °C), S-D (coal heated at 300 °C), S-E (coal heated at 400 °C), S-F (coal heated at 500 °C), S-G (coal heated at 600 °C), S-H (coal heated at 700 °C), S-I (coal heated at 800 °C), S-J (coal heated at 900 °C), S-K (coal heated at 1000 °C), S-L (coal heated at 1100 °C), S-M (coal heated at 1200 °C), respectively.

2.3. Experimental analysis

Proximate analysis (moisture, ash yield, volatile matter and fixed carbon) was performed on the raw coal sample according to Chinese coal standard GB/T 212-2008, which is comparable to ASTM standards (ASTM-D3173-11, 2011; ASTM-D3174-11, 2011; ASTM-D3175-11, 2011) except for small temperature difference used in the measurement procedures. Ultimate analysis (C, H, N) was performed by a vario MACRO cube Elemental Analyzer, and the O content was calculated by difference. The total sulfur and various forms of sulfur was measured by WS-S101 Automatic Sulfur analyzer, according to Chinese standard GB/T 214-2007 (equivalent to ASTM standard D3177-02, 2002) and GB/T 215-2003 (equivalent to ASTM standard D2492-02, 2012), respectively.

The SDT Q600 simultaneous thermal analyzer was used for the thermogravimetric analysis. About 20 mg of raw coal sample was weighed for each analysis in an air or N_2 flow of 100 mL/min. The analysis were conducted in a temperature range of 20 °C–1200 °C, with a constant linear heating rate of 10 °C/min. The weight loss and the rate of weight loss were monitored continuously. The X-ray polycrystalline diffractometer (XRD, 9KW/SmartLab, Japan) with Cu-Kα radiation was used to identify the crystalline phases of raw coal and twelve heated coal samples. The diffraction patterns were recorded over a 2θ interval of 5–70°, with a step increment of 0.01°. The minerals in each sample were identified by comparison to the mineral phases in the International Centre for Diffraction Data (ICDD) power diffraction file (PDF) database. Photo-micrographs of the heated coals produced at 900 and 1200 °C were obtained using a scanning electron microscope (qutanta 400 FEG SEM), equipped with an energy-dispersive microanalyser (EDAX). Similar SEM analytical procedure were previously reported (Saikia et al., 2015; Tezza et al., 2015; Wilcox et al., 2015; Dalmora et al., 2016; Ramos et al., 2017). Fourier transform infrared spectroscopy (FTIR) analysis was used to determine the functional group of raw coal and heated coal at 1200 °C (i.e. high temperature coal ash. Analysis was recorded in the 4000–400 cm^{-1}) frequency region using a Thermo Nicolet 8700 spectrometer through KBr pellets.

Approximately, 0.1 g of raw coal and heated coal samples were digested using HCl-HNO_3-HF (3:3:2) in a microwave reactor. The total concentrations of Co, Cr, Ni, and V were analyzed by inductively coupled plasma atomic emission spectrometry (ICP-AES). The accuracy of elements was evaluated using the standard reference material SARM20 (coal) and GBW07406 (GSS-6) (soil). The precision of these elements is within ±5%.

3. Results and discussion

3.1. Proximate and ultimate analysis

Proximate and ultimate analysis of experimental raw coal were listed in Table 1. The raw coal contained 0.94% moisture, 42.74% ash yield, 17.07% volatile matter, and 39.25% fixed carbon. The C, H, O, N, S and calorific value were 54.82%, 3.94%, 6.5%, 0.98%, 0.84% and 12.68 MJ/kg, respectively. Pyritic sulfur was the primary form, followed by organic sulfur and sulfate sulfur. These data indicated that the raw coal was characterized by high ash yield, and low moisture, volatile matter, sulfur and calorific values according to Chinese coal classification standards (MT/T 850-2000, GB/T 15224.1, MT/T 849, GB/T 15224.2 and GB/T 15224.3).

3.2. TG, DTA and DSC characterization

Thermogravimetry (TG), differential thermal analysis (DTA) and differential scanning calorimetry (DSC) have been widely applied for quantitative characterization of coal combustion behavior and mineral alternation (Iordanidis et al., 2001; Zhou et al., 2012). The experimental TG, derivative TG (DTG), DTA and DSC curves of the raw coal sample under the flows of air and N_2 were illustrated in Fig. 1. According to TG curve (a) under the air flow, there were four stages during combustion of the studied coal sample. The stage of

Table 1
Proximate and ultimate analysis of raw coal.

	raw coal
M_{ad} (wt.%)	0.94
A_{ad} (wt.%)	42.74
V_{ad} (wt.%)	17.07
FC_{ad} (wt.%)	39.25
Low heating value (MJ/kg)	12.68
C (wt.%)	54.82
H (wt.%)	3.94
O (wt.%)	6.5
N (wt.%)	0.98
S_t (wt.%)	0.84
S_p (wt.%)	0.51
S_s (wt.%)	0.01
S_o (wt.%)	0.32

ad: air dry basis; M: moisture; A: ash yield; V: volatile matter; FC: fixed carbon; S_t: total sulfur; S_p: pyritic sulfur; S_s: sulfate sulfur; S_o: organic sulfur.