



Full Length Article

The retention mechanism, transformation behavior and environmental implication of trace element during co-combustion coal gangue with soybean stalk



Chuncaï Zhou ^{a,b}, Guijian Liu ^{a,b,*}, Zhongyu Xu ^a, Hao Sun ^a, Paul Kwan Sing Lam ^c

^a CAS Key Laboratory of Crust-Mantle Materials and the Environments, School of Earth and Space Sciences, University of Science and Technology of China, Hefei 230026, China

^b State Key Laboratory of Loess and Quaternary Geology, Institute of Earth Environment, The Chinese Academy of Sciences, Xi'an, Shaanxi 710075, China

^c State Key Laboratory in Marine Pollution and Department of Biology and Chemistry, City University of Hong Kong, Tat Chee Avenue, Kowloon, Hong Kong, China

HIGHLIGHTS

- The gaseous trace elements were reduced during co-combustion coal gangue with biomass.
- The transformation behaviors of trace elements were attributed to their associations.
- The environmental implications of trace elements in co-combustion residues should be concern.

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ABSTRACT

A laboratory-scale circulating fluidized bed combustion experiment was performed to investigate the retention mechanism and transformation behavior of trace elements (TEs) during co-combustion of coal gangue (CG) with soybean stalk (SS). Chemical sequential extraction was conducted to determine the associations of the TEs. Results show that the distribution and transformation behavior of the selected TEs during co-combustion are variable, which is attributed to the different physico-chemical properties, associations of the TEs and ash components. The elevated retention of gaseous TEs during co-combustion may be explained by the surface-reactions with active minerals, chemisorption, and the interactions with ash components and elements. The gaseous phase, exchangeable and residual fractions are increased with the decomposition of the Fe-Mn oxides fraction in the oxidizing atmosphere. The combustion ash may pose a medium environmental risk if no countermeasure is adopted.

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1. Introduction

Coal gangue (CG) is a significant residue of coal mining activities (accounting for approximately 10–15% of raw coal production), and is characterized as having a high ash yield, low carbonaceous content and low heating value [1]. Because of the excellent mass and heat transfer properties of circulating fluidized bed combustion technology, the utilization of CG for electricity production has increased steadily in both capacity and quantity [2,3]. Coal combustion is regarded as a significant source of atmospheric pollutants [4]. Besides the conventional pollutants (SO₂, NO_x, particu-

late matter and organic compounds) of environmental concern, the emissions of TEs may have severe environmental impacts and are of increasing concern [5]. Recent research into the nature of emissions of TEs from coal-fired plants has reported that TEs may be associated with particulate matter in physical and/or chemical forms and constitute a significant fraction of the environmental implication of the emissions from coal combustion [6]. The adverse effects associated with high emissions of TEs include acute and chronic lung injuries, inflammation, altered sensitivity of the epithelia, changed immunological mechanisms, and increased cancer risk [7]. The elevated concentrations of TEs are found in CG when compared with that of coal [8,9]. Therefore, TE control and management during CG firing is extremely importance.

Much effort has been expended in studying the transformation mechanisms of TEs during coal combustion, and published data indicate that the redistribution behavior of TEs is greatly deter-

* Corresponding author at: CAS Key Laboratory of Crust-Mantle Materials and the Environments, School of Earth and Space Sciences, University of Science and Technology of China, Hefei 230026, China.

E-mail address: lgj@ustc.edu.cn (G. Liu).

mined by the solid/gas reactions in the combustion system, feedstock physico-chemical properties, volatility and associations of TEs in feedstock, and combustion conditions [6,10]. Intensive studies have addressed the interactions between TEs with the ash components (Ca, Fe, and Al) and reported that these interactions play an important role in the partitioning behavior of TEs in the gas or solid phase [11]. Based upon these interactions, a large quantity of sorbents, including limestone, kaolinite, aluminum oxide, apatite have been tested during coal combustion in fixed bed, drop tube furnaces, and fluidized bed boilers [12]. Among these sorbents, the Ca-based sorbents were regarded as the effective and available materials for the retention of gaseous TEs during coal combustion [13]. The adsorption mechanisms of Ca-based sorbents may be attributed to chemical reaction and physisorption [14]. As a high alkali and alkali-earth (Ca, K, Na, Mg) containing material, biomass was employed to capture TEs in our previous study, which showed that the emissions of TEs were reduced [15]. In this study, a further effort is made to elucidate the TE retention mechanism and transformation behavior during co-combustion of CG with biomass.

Generally, a suitable proportion of high value coal is added into the current CG combustion boiler to promote the ignition performances and thermal reactivity. Biomass contains more volatile matter (and thus has higher thermochemical reactivity), lower ignition temperature and shorter combustion duration in comparison to fossil fuels [16–18]. Our previous study suggested that biomass could be used as substitute of high value coal in the CG combustion and provide a stable flame, enhance the ignition and thermal behavior during co-combustion [19,20]. Meanwhile, biomass is considered to be a clean and readily available renewable and cost effective energy source for CO₂ neutrality [21]. Therefore, the co-combustion of coal gangue with biomass could not only relieve the rapid consumption of finite fossil fuels and the overload of solid wastes, but also reduce the emissions of TEs and environmental pollution. Based on the aforementioned advantages, it is crucial to carry out a systemic investigation on the retention mechanisms and transformation behavior of TEs during co-combustion of coal gangue with biomass to provide an alternative for TE control and management.

Soybean stalk (SS) is a typical, abundant and cost-effective agriculture biomass residue in China, which contains a high content of alkali and alkali-earth elements (the content of Ca > 30 wt.%) [15,16]. Thus, the SS was selected in this study. The main objectives of the present study were to determine (1) the relative distribution characteristics of TEs, (2) the retention mechanism and transformation behavior of TEs during co-combustion CG and SS, (3) potential environmental implication of co-combustion residues.

2. Materials and methods

2.1. Materials

CG samples were obtained from the floor of No. 7 coal seam of Guqiao Coal Mine, Huainan, China, using the cutting channels downwards method. The selected samples were regarded as being a representative sample of the coal seam. The SS, known to be an abundantly available agricultural biomass in the study area, was collected at the farmland. After being collected, these samples were immediately placed in sealed plastic bags to avoid contamination. Both the selected CG and SS were air-dried, ground and sieved to pass through 2 mm mesh for subsequent analysis. The proximate, ultimate and trace element analyses of the selected samples were published in the previous study [20] and are summarized in Table 1. In comparison with CG, the obvious differences

related to the composition and properties of SS include both elevated contents of moisture, volatile matter, H, O, alkaline-earth elements and low values of C, fixed carbon, ash yield, N, S, Si, ignition temperature and the TEs. The PHILIPS X' Pert PRO X-ray diffractometer (XRD) with Cu K-alpha radiation were employed for the mineralogical analysis. The pattern was recorded over a 2θ interval of 10–60°, with a step increment of 0.01°. The minerals were compared and identified by reference to the ICDD powder Diffraction File.

2.2. Combustion procedure

The thermochemical and combustion behavior of CG during co-combustion with SS were published in a previous paper [20], the results suggesting that the blending ratio of 20–30% SS content is regarded as the optimal proportion for blending and could be combusted in the current combustion system directly with few modifications. Therefore, the CG/SS blend sample at a weight ratio of 80:20 was employed in the subsequent analysis.

A thorough description of the experimental equipment employed can be found in a previous study [15]. Briefly, the mixed fuel was pneumatically placed in a fluidized bed reactor, equipped with an agitation chamber and an insulated injector to obtain the uniform fuel feeding rate. The combustion temperature of the boiler was regulated by a batch of electrical resistances, and was set at 900 °C in this study. The feeding rate, the excess air ratio (relative to the theoretical air), the fluidizing air flow, the secondary air flow and the fuel/air ratio are 0.2 kg/h, 40%, 1 Nm³/h, 0.5 Nm³/h and 0.714, respectively. The feeding fuel, bottom ash and fly ash were sampled at each site, and crushed to pass through a 200-mesh sieve for the subsequent analysis. The individual fuel (CG and SS) and their blending sample (FS) were analyzed in triplicate to ensure the repeatability and representativity.

2.3. Chemical analysis

TE (As, Cd, Cu, Cr, Ni, Pb, V, and Zn) analyses of individual fuel, mixed fuel and their ash samples were determined by using inductively coupled plasma mass spectrometry (ICP-MS). Detail of the TE analysis procedures, particularly the chemical digestion methods, can be found elsewhere [22,23]. The selected samples and standard reference materials (NIST 1632b (coal) and GBW07406 (soil)) were initially digested by an acidic mixed solution (HCl:HNO₃:HF = 3:3:2) in a microwave oven. The analytical method was calibrated by the standard reference materials. Recoveries of 96.2–104% were obtained for all the selected materials. Each sample analysis was conducted in triplicate. The acceptable precision was within ±5% for all the TEs.

2.4. Sequential chemical extraction procedure

In order to investigate the transformation behavior of TE during co-combustion, the TEs in the individual fuel, blending fuel and their corresponding combustion residues were extracted and classified into five phases by the empirical sequential chemical extraction procedure described by Tessier et al. [24]. The TEs could be partitioned into five fractions including exchangeable fraction (F1), carbonates bound fraction (F2), Fe-Mn oxides bound fraction (F3), organic matter bound fraction (F4), and residual fraction (F5). The TEs in each fraction were analyzed by ICP-MS.

2.5. Leaching tests

In order to evaluate the potential mobility and environmental implication of TEs in co-combustion residues, the compliance leaching test EN 12457 Part 2 procedure was conducted [25].

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