



## Full Length Article

# A comparative study on the mineralogy, chemical speciation, and combustion behavior of toxic elements of coal beneficiation products



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## ABSTRACT

The huge demand for high-quality coal in China has resulted in increased generation of preparation plant wastes of various properties. A series of beneficiation products collected from a preparation plant were characterized to understand their petrographic and mineralogical characteristics, as well as thermochemical and trace element behavior during combustion. The minerals in the Luling preparation plant wastes from Huaibei coalfield mainly included kaolinite and quartz, with minor calcite, ankerite, pyrite, illite, chalcopyrite, albite, K-feldspar, anatase/rutile, and iron-oxide minerals. Massive clay lumps of terrigenous origin, cleat-infilling carbonate, and pyrite of epigenetic origin were prone to be enriched in the middlings and coal gangue. Minor or trace heavy minerals also reported to the preparation plant wastes. The contents of low-density density vitrinite and liptinite were enhanced in the clean coal, while inertinite-maceral group were enriched in the middlings. The modes of occurrences of toxic elements differed between raw coal and the waste products; and their transformation behavior during heavy medium separation is largely controlled by clay minerals (V, Cr, Co, Sb, and Pb), carbonate minerals (Co and Pb), sulfide minerals (As, Cu, Ni, Cd, and Zn) and organic matters (V, Cr, Se, and Cu). Three groups were classified based on the volatile ratio (Vr) of toxic elements. Group 1 includes the highly volatile element Se with Vr > 85%; Group 2 contained elements As, Pb, Zn, Cd and Sb, with the Vr in the range of 20–85% and V, Cr, Co, Ni and Cu with Vr less than 20% were placed into Group 3. Thermal reactivity of coal inferred from the combustion profiles could be significantly improved after coal beneficiation, whereas the increased inorganic components probably inhibited the thermal chemical reaction of wastes.

## 1. Introduction

To upgrade the quality of coal for industrial utilization, the ratio of cleaned coal to raw coal in China has rapidly increased in the past decades. According to statistics up to 2011, the total amount of preparation plant rejects is more than 3 Gt in China. It has been well-documented that toxic trace elements in coal were prone to be associated with mineral matter, especially for high-ash coal with large amounts of heavy-metal-bearing minerals such as pyrite, barite, rutile/anatase, among others [1–5]. Minerals such as clay and pyrite in coal are targeted for removal in coal preparation to reduce the sulfur, mineral matter, and associated toxic trace elements [6–9]. Different types of coal preparation plant rejects show wide variations in physiochemical properties, such as ash yield, moisture content, maceral

distribution, mineral compositions, etc. [9–11]. Recently, much attention has been paid to the research and application of three-product dense medium cyclones (DMCs) in China [12,13]. The waste product types produced from the DMCs (Fig. 1) include high-ash coal gangue, fine middling coal, and coal slime of ultrafine coal with high water contents. However, there are only a few works focusing on the petrologic, mineralogical and geochemical characteristics of different coal beneficiation products generated from DMCs units in coal preparation plants [11,14]. In addition, while correlations between element partitioning and physical separations are usually used as the basis to infer the elemental associations [9,10], data on the chemical speciation of trace elements in individual beneficiation products are still lacking.

On the other hand, the cleaning wastes contain combustible materials which could be used in electricity generation and are regarded as a

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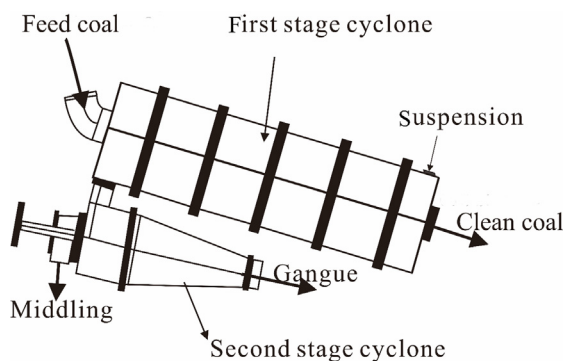


Fig. 1. Sketch map of three-product DMCs (3GDMC1200/850A) in Luling preparation plant, based on the data provided by the local technicians.

potentially valuable energy resource [15–18]. Some countries such as Australia [17,18], India [19], and China [20] have developed fluidized-bed combustion (FBC) technology as a means for recovering energy from preparation plant wastes. Thermogravimetric analysis (TGA) has been extensively used in studying the reactivities of carbonaceous materials to assess the thermochemical properties of coal, biomass, and so on [21,22]. The combustion profile of TGA depends upon various parameters such as coal rank, maceral distribution, and mineral contents. In addition, due to the enrichment of some toxic elements in the high-ash waste products, the volatile characteristics of the elements during preparation plant wastes combustion could be of concern. The volatility of toxic trace elements is mainly a function of modes of elemental occurrences and combustion conditions [23,24,26,28–30]. Differences in maceral composition, coal rank, and mineral matter of preparation plant wastes may result in different combustion reactivities and volatile tendencies of toxic elements [25,26].

The objectives of this study were to use multidisciplinary means to investigate (a) the petrology, mineralogy, and geochemistry of various preparation plant wastes produced from DMC, (b) thermal characteristics of cleaning wastes via thermal gravimetric analysis, and (c) volatile characteristics of toxic elements during coal cleaning waste combustion. The results of this study are expected to provide a useful basis for treating the cleaning waste products in an environmentally friendly and economical manner.

## 2. Materials and methods

### 2.1. Background and sample collection

In this study, a series of samples including raw coal, middlings, clean coal, slime, and coal gangue were sequentially collected from the DMCs of Luling coal preparation plant in the Huaibei coalfield (Fig. 1). The feed coal in the plant all came from the coal seams 8, 9 and 10 of Luling mine of Huaibei coalfield. The heavy dense medium used in the DMC unit was mainly composed of magnetite powder. As shown in Fig. 1, during the cleaning process, the feed consisted of blended coal and dense medium that enters tangentially near the top of the cylindrical section of the cyclone can form a strong swirling flow. Due to the centrifugal forces, heavy fractions of parent coal with high specific gravity should move towards the wall and discharge in the underflow through the spigot, while light fractions could be caught in an upward stream and generated clean coal from the overflow outlet [27]. The suspended liquid that contained 0.25–2.0-mm coal particles generated from the overflow will enter into the slime dense medium cyclones, which can recover the clean coal and produce tailings. Coal slurry produced from the whole process of DMC will enter into the flotation cell to recover the clean coal, and the tailing slurry will be concentrated and then dewatered by centrifuges and filter press. The resulting three cleaning products (clean coal, middlings, and gangue) were screened to

recover the medium and yielded the final products. DMCs in the Luling preparation plant can process raw coal at 3 Mt/a. The gangue, middlings, and slime were generally used for generating electricity in the local power plants [28], occasionally the coal gangue was used as a raw material in a brick-making plant [29].

All the samples were homogenized and reduced to obtain a representative 2–3 kg sample, and then were immediately sealed into plastic bags to avoid possible oxidation and contamination. Each sample was dried at 40 °C for 24 h, and then crushed in an agate mortar to pass through the 60-mesh sieve for different experiments.

### 2.2. Analytical methods

The contents of C, H, and N in the sample were determined using an elemental analyzer (Vario EL cube). Proximate analysis (ash yield, moisture, and volatile matter) was analyzed based on ASTM Standards D3174-04, D3173-04 and D3175-04 (ASTM, 2007), respectively. Total sulfur and forms of sulfur were determined following ASTM Standards D3177-02 and D2492-02 (ASTM, 2007), respectively. Petrographic analysis of all samples was performed at the University of Kentucky Center for Applied Energy Research. Each sample was ground to less than 1 mm and prepared as epoxy-bound pellet and examined with 50× oil-immersion reflected-light optics using Leitz Orthoplan microscopes. The mineralogical phases in the waste products were determined by X-ray powder diffraction, supplemented by examination using a field emission scanning electron microscope (TESCAN MIRA3 LMH Schottky FE-SEM), equipped with an energy dispersive analysis system of X-ray (EDAX, Genesis APEX Apollo System). The XRD analysis of each sample was carried out on a Philips X'Pert PRO X-ray powder diffraction with Cu K-alpha radiation, and the patterns are recorded over a 2θ interval of 3–70°, with a step increment of 0.02°. The working distance of the SEM was about 10 mm with beam voltage 15 or 20.0 kV. Samples were carbon coated and then were mounted on aluminum SEM stubs using sticky conductive tapes.

### 2.3. Chemical analysis

The major element oxides were determined by XRF (XRF-1800). Before XRF analysis, all the samples were ashed at a temperature of 815 °C, an alkaline fusion method for the high temperature ash were employed prior to the XRF analysis. The loss-on-ignition (LOI) was also determined at 815 °C [31]. Trace elements in the samples were determined by ICP-MS (Thermo Fisher, X Series II), and As and Se were determined using AFS (AFS-230Q). Approximately 0.05 g of each powder sample (200-mesh) was transferred into a PTFE digestion vessel with 2-ml purified HNO<sub>3</sub> overnight, and then added to an acid mixture (HNO<sub>3</sub>: HF = 2:5) in a programmed microwave oven (from room temperature to 120 °C in 10 min and kept for 10 min; then increased to 160 °C in 5 min and kept for 10 min; finally increase to 210 °C and kept for 60 min). Finally, each solution was filtered through a 0.45-μm membrane and made up to 25-ml with Milli-Q water with 3% HNO<sub>3</sub>. Two percent HNO<sub>3</sub> solution was injected into the ICP-MS system to eliminate the memory effect of the previous sample. The Re internal standard solution was used for tuning and checking of the ICP-MS calibration, obtaining a RSD lower than 8%. Blanks, certified reference materials SARM-20 (coal) and GBW07406 (soil) were digested and determined following the same procedure used for checking the accuracy of the trace elements. Analytical errors were less than 5% for most elements and around 10% for V (108.3%), and Pb (106.2%).

A six-step sequential chemical extraction (SCE) was used to determine the modes of occurrences of trace elements in the coal waste samples. The SCE procedural separates the trace elements into six fractions: water-leachable, ion-exchangeable, carbonate-bound, organic-bound, silicate-bound, and sulfide-bound. The detailed SCE procedure for toxic elements (V, Cr, Co, Ni, Cu, Zn, As, Cd, Se, Pb, and Sb) of environmental concern in the samples was based on Dai et al. [32].

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