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Occurrence and characteristics of abundant fine included mineral particles in Collie coal of Western Australia

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ABSTRACT

Collie coal is the only coal being mined in Western Australia and plays an important role in supplying cheap energy to the State's energy mix. This study reports a systematic investigation into the occurrence and characteristics of the fine mineral matter in Collie coal that is crushed to the size of $< 212 \mu\text{m}$ and density separated. The results show that 94.1 wt% of the mineral matter in the whole Collie coal is present in the three density fractions of 1.2–1.4, 1.4–1.6 and ≥ 2.0 , including 26.8 and 58.6 wt% as included mineral matter in two density fractions with specific gravities of 1.2–1.4 and 1.4–1.6, and 8.7 wt% as excluded mineral matter in the density fraction of ≥ 2.0 . Further analysis of these key Collie coal density fractions using computer-controlled scanning electron microscopy (CCSEM) shows that 54.13 wt% of the total mineral matter in the whole Collie coal are fine mineral particles of sizes $< 10 \mu\text{m}$. These included fine mineral particles ($< 10 \mu\text{m}$) are distributed as 20%:80% in the two coal density fractions of 1.2–1.4 and 1.4–1.6, which contribute to 10.64 and 43.49 wt% of the total mineral matter in the whole coal, respectively. CCSEM analysis also shows that the key minerals in the fine mineral particles are quartz, kaolinite, K-, Ca- or Fe-Al silicates and pyrite-related minerals. However, large proportions of these included fine mineral particles have “unclassified” mineral phases. Such unclassified particles contribute to 43.88 and 36.84 wt% of the fine mineral particles with size $< 10 \mu\text{m}$ present in the coal density fractions of 1.2–1.4 and 1.4–1.6 (equivalent to 4.67 and 16.02 wt% of the total mineral matter in the whole coal), respectively. The chemistry data show that these individual fine mineral particles ($< 10 \mu\text{m}$) contain mixed compositions of Si + Al, Fe + Ti and other elements such as Ca, K, S and P, suggesting that one single fine mineral particle contains multiple mineral phases.

1. Introduction

Mineral matter in coal is an important consideration for coal in various thermochemical utilisation processes (e.g. combustion, gasification and ironmaking) [1–6]. It experiences a series of physical and chemical transformation and leads to various notorious ash-related issues including slagging, deposition and fouling [1–4,7,8] and particulate matter emissions [9–14]. Transformation of mineral matter during coal thermochemical processing is influenced by two key factors. One is the properties of mineral matter in coal. For example, excluded mineral particles experience respective thermochemical transformation depending on mineralogy while included mineral matter in reacting coal particles experiences different temperature-time history and local

reaction atmosphere [1]. Being associated with other included mineral matter within the same coal particle, included mineral matter may experience coalescence during coal reactions [1,11,15–17]. Depending on the original mineral compositions, coalescence of dissimilar minerals within the same coal particles can result in the formation of lower melting eutectics, increasing ash particle size and ash deposition propensity [1,11,15]. The other key factor is the thermochemical processing conditions [3,18–20]. For example, in a boiler, ash slagging and fouling are strongly dependent on local combustion conditions, varying throughout different sections of the boiler with ash deposits of different characteristics [18].

In Western Australia (WA), Collie coal is the only coal currently being mined in the state. It is an important cheap energy source for

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power generation in stationary coal-fired power stations. Over the years, there have been scattered studies on the properties of Collie coal and the reactions of coal organic and inorganic matter during thermochemical processing [18,21–26]. One interesting observation is that abundant fine ash particles are produced from Collie coal combustion, playing important roles in ash formation, deposit initiation and growth in the boilers of WA coal-fired power stations burning pulverised Collie coal [18,22]. Fine ash particles can be formed from large mineral particles in coal via various mechanisms such as thermal shock, fragmentation and chemical reactions [1,15]. The original fine mineral particles inherent in coal particles can also be an important source for the formation of fine ash particles and PM₁₀ emission during pulverised coal combustion [26]. Properties including particle size distribution (PSD) and mineral compositions of inherent fine mineral matter in coal may be an important consideration in coal selection guideline for practical applications.

Therefore, this study aims to carry out a systematic investigation into the occurrence and characteristics of fine mineral particles in Collie coal that plays an important role in WA's energy mix. The program considers an array of experimental and analytical techniques including density separation, ash chemistry analysis, scanning electron microscope (SEM), and computer-controlled scanning electron microscope (CCSEM). CCSEM is an advanced automated technique for characterising mineral particles in coal on a particle-by-particle basis, which was initially developed in late 1970s [27,28] and widely used since 1990s [2,29–33]. It offers the advantage of acquiring comprehensive data on the size distribution, composition and associations of mineral particles in coal.

2. Experimental section

2.1. Coal sample preparation

Collie coal sample was collected from Collie power station, Collie, Western Australia. The coal sample was crushed and pulverised to a size of < 212 μm which is hereafter referred to as “raw coal”. The raw coal was then separated via sink-float method into a series of specific gravity cut-offs of 1.2, 1.4, 1.6, 1.8 and 2.0, respectively. A total of six density-separated coal samples were prepared for subsequent experiments, including coal density fractions of (a) “< 1.2”, i.e. coal sample with specific gravity < 1.2; (b) “1.2–1.4”, i.e. coal sample with specific gravity ≥ 1.2 but < 1.4; (c) “1.4–1.6”, i.e. coal sample with specific gravity ≥ 1.4 but < 1.6; (d) “1.6–1.8”, i.e. coal sample with specific gravity ≥ 1.6 but < 1.8; (e) “1.8–2.0”, i.e. coal sample with specific gravity ≥ 1.8 but < 2.0; and (f) “ ≥ 2.0 ”, i.e. coal sample of specific gravity ≥ 2.0 , respectively.

2.2. Characterisation of samples

2.2.1. Proximate and ultimate analyses

The proximate analysis of coal samples was carried out using a thermogravimetric analyser (TGA, model: METTLER) according to ASTM D3172. The ultimate analysis was conducted using a combination of several methods. The contents of carbon (C), hydrogen (H) and nitrogen (N) in a sample was determined using an elemental analyser (model: Perkin Elmer 2400 Series II), following the procedure detailed in AS1038.6.4. The content of S in the samples was determined using improved Eschka method [34]. The oxygen (O) content was determined by difference on a dry-ash-free (daf) basis.

2.2.2. Ash chemistry analysis

Ash chemistry analysis of coal samples was carried out following a previous method based on inductively coupled plasma-optical emission spectroscopy (ICP-OES) [35]. The method consists of three steps: ashing, fusion, and subsequent analysis using an ICP-OES (model: PerkinElmer Optima 8300). Briefly, a coal sample was firstly ashed in a

platinum (Pt) crucible under O₂ atmosphere according to a designed temperature-time program that prevents the loss of ash species during coal oxidation. At an ash to flux mass ratio of 1:30, the ash sample in the Pt crucible was then mixed with the X-ray flux (35.3% lithium tetraborate and 64.7% lithium metaborate), followed by fusion at 950 °C for 2 h. The fusion bead was dissolved in dilute nitric acid (5 wt %) and subjected to the ICP-OES for ash chemistry analysis.

2.2.3. Scanning electron microscope (SEM) characterisation

Cross-section characterisation for coal samples were carried out using a SEM (model: Philips XL30). Briefly, a sample pellet was prepared via mixing a coal sample with epoxy resin that was then cross-sectioned and polished. The pellet was then coated with carbon prior to SEM characterisation.

2.2.4. Thermogravimetric (TG) analysis

The Collie coal with a density fraction of ≥ 2.0 was also subjected to TG analysis. Briefly, ~ 10 mg coal sample was heated to and held at 110 °C for 30 min to remove moisture, and then heated to and held at 800 °C for 20 min, under either air or argon (flow rate: 100 mL/min). The heating rate of all the temperature ramping processes was 10 °C/min.

2.2.5. Computer-controlled scanning electron microscope (CCSEM) analysis

The CCSEM (consisting of a JEOL JSM5600 scanning electron microscope and a CDU-LEAP energy dispersive X-ray spectrometer) located at Chubu University was employed for the analysis of the mineral matter in Collie coal samples, following the procedure detailed in a previous study [33]. Briefly, a coal sample was set into resin, cross-sectioned, polished and then carbon coated to prepare a sample pellet for CCSEM analysis. Based on the difference in the intensity of the backscattered electron signals, coal and mineral matter could then be detected. For each mineral particle, the X-ray spectra were acquired for 12 s of live time at an accelerating voltage of 20 kV for each mineral particle, with the relative dead times of the EDX spectrometer being < 25%. Minerals in coals were determined based on the elemental composition of individual particles according to the composition criteria developed previously [36]. The CCSEM analysis provides data on the PSD of mineral particles, together with the chemical composition of each mineral particle in the coal.

3. Results and discussion

3.1. Distribution of coal mass and mineral matter in various Collie coal density fractions

Fig. 1a presents the data on coal mass distributions across the series of density fractions of the Collie coal. It is interesting to see that after density separation, the vast majority (97.2 wt%) of coal mass is present in the two density fractions, i.e. 1.2–1.4 and 1.4–1.6, which constitute to 29.5 wt% and 67.7 wt% of the total coal mass (on a dry basis), respectively. There are only small amounts of coal mass in the density fractions of < 1.2, 1.6–1.8, 1.8–2.0 and ≥ 2.0 , i.e. merely 0.2, 1.7, 0.2 and 0.7 wt% of the total coal mass, respectively.

Fig. 1b illustrates the ash contents of all the density fractions. It can be seen that the ash contents of the density fractions of < 1.2, 1.2–1.4 and 1.4–1.6, 1.6–1.8, 1.8–2.0 and ≥ 2.0 are 10.1, 6.3, 6.0, 16.7, 51.2 and 92.8 wt% (on a dry basis), respectively. There is a general trend that the ash contents in coal samples increase with the density of coal samples. This is as expected since mineral matter has a considerably higher specific density (> 2.0 [1]) than that of coal organic matter (typically ~ 1.2 [37]). However, it is noted that the ash content of the density fraction of < 1.2 is actually higher than those of the two heavier density fractions of 1.2–1.4 and 1.4–1.6, suggesting that the density of organic matter in the density fraction of < 1.2 is lower than those of

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