



Size effects on the thermal behavior of superfine pulverized coal ash



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ABSTRACT

The coal ash fusibility is an important parameter that affects the boiler design and operation significantly. Superfine pulverized coal is beneficial to the alleviation of fouling and slagging problems due to its distinctive properties in the aspects of multiphase flow, heat transfer and combustion atmosphere. In this work, the size related ash fusion propensity were emphasized from the point of view of the chemical and mineralogical morphology of coal ash. A comprehensive study on the chemistry and thermal behavior of superfine pulverized coal ash was conducted combining multiple analytical methods such as XRF (X-ray fluorescence), XRD (X-ray diffraction), TGA (thermo-gravimetric analysis), slagging indices, AFT (ash fusion temperatures) tests and phase diagrams. Besides, a quantitative analysis method was proposed to eliminate the subjective judgments in AFT tests through a numerical image-processing platform, which could depict a complete deformation profile of the whole ash fusion process with high precision and reproducibility. The results from this work are helpful for understanding the size dependence of ash thermal behavior, and predicting the ash fusion propensity in the future application of superfine pulverized coal.

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1. Introduction

The ash thermal behavior plays a crucial role in the boiler design and operation [1]. The deposition of non-combustible material in coal ash induces serious slagging and fouling problems that have significant impacts on the performance of power utilities such as the efficiency, economy, safety, and reliability [2]. Besides, the ash fusibility is an important parameter to be monitored and optimized during coal utilization like combustion, liquefaction, gasification, and ash reuse [3–5]. The adverse effects of ash deposits on the thermal conductivity and emissivity have been widely discussed [2], with the corrosion and erosion problems being reviewed [6–9]. In recent years, there is an increasing demand for the use of renewable resources [10], considering the sustainable development of energy and environment. Therefore, combustion and co-combustion of biomass that is one of the most widely used renewable energy has drawn more and more attention. However, the abundance of inorganic material in biomass, especially alkali metals, aggravates the ash fusion problems. Besides, the strength and tenacity of biomass deposition are higher due to the existence of certain crystal skeleton components with high melting points (e.g., $\text{CaMg}_3\text{O}(\text{Si}_3\text{O}_9)$) [10]. Thus, additional ash removing

efforts may be required during co-combustion process [11]. Therefore, it is of great concern to determine the deposition propensity of the ash.

On the other hand, the serious global warming problems induced by emission of anthropogenic greenhouse gases from fossil fuel combustion have gained increasing concern [12]. A variety of carbon capture and sequestration (CCS) methods has been proposed, of which the oxy-fuel combustion technology is considered as the most promising option in the near future [13,14]. However, the reductive atmosphere in O_2/CO_2 combustion lowers the fusion points of coal ash, leading to more serious fouling issues and corrosion phenomena. Interestingly, it was observed during our previous work that superfine pulverized coal played an important role in the alleviation of ash and slag buildup in a boiler. We explained this phenomenon from the particular point of view of flow, combustion atmosphere, and heat transfer. With the increase of particle sizes, the inertial impaction leads to the buildup of coarse-grained deposit with elliptical or mountain-like shapes [7], which aggravate the slagging and fouling problems. On the other hand, the diffusion and thermophoresis are the dominant effects during the transport of fine particles, which only produces a thin layer of evenly distributed deposition [2,7]. Therefore, larger ash particles are more susceptible to deposit on the tubes [6]. Furthermore, the coal particle sizes have significant influence on the combustion atmosphere. Larger coal particles consume more oxygen, elevate

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the temperatures and thus result in a reductive local environment near the furnace walls, which aggravates the ash fusion propensity. In this work, the superiority of the superfine pulverized coal on the ash fusion propensity was interpreted from the aspects of the chemical and mineralogical morphology and the size related thermal behavior of coal ash.

The ash fusion temperatures (AFT) and empirical indices that derived from chemical properties have been extensively applied to predict the ash deposition behavior [15–19]. The coal ash chemistry is always expressed as the percentage of several fundamental oxides that are frequently detected [20]. The slagging indices as an easy and expeditious methodology deduced from the correlations of these oxides are still widely used in laboratory research and practical application despite of certain deficiencies. For example, the laboratory prepared ash in the tests cannot simulate the ash forming under the real combustion or gasification conditions [21]. In addition, these indices may not be applied universally due to the complexity of coal ash mineralogy [22]. However, the empirical relationships can obtain good results in predicting ash deposition propensity with similar chemical composition. Furthermore, the deviation can be partly compensated through the consistent settings of test conditions [16]. Therefore, certain conventional slagging indices such as base-to-acid ratio (B/A) and silica ratio (G) can still be instructive, which were adopted here to study the size effects on the ash fusion behavior.

The AFT analysis as an inexpensive and standardized method has also been widely employed in the ash fusion research. Although various criteria have been adopted in eight different international procedures (e.g., ISO, ASMT, BS, and PRC, etc.) [20], typically four characteristic temperatures are recognized, i.e., the deformation temperature (DT), softening temperature (ST), hemispherical temperature (HT) and flow temperature (FT). The major concern against AFT tests is the subjectivity and reproducibility. The results may be misleading due to the reliance on the visual observation of individual operators. The deviation can reach 30 °C or even higher. Therefore, an improved temperature-determination procedure was proposed here to fix the drawbacks of the conventional method through an image processing platform. The pictures recorded from AFT tests were numerically treated, and abundant geometrical information was extracted. The quantitative analysis of images could eliminate the subjective judgments, and make the results be more convincing and reproducible.

Above all, superfine pulverized coal is beneficial to the alleviation of fouling and slagging problems in utility boilers. However, there is little knowledge about the size effects on the chemical and mineralogical morphology of coal ash, and the size related ash fusion propensity should be emphasized. In this paper, a comprehensive study on the chemistry and thermal behavior of superfine pulverized coal ash was conducted combining multiple analytical methods such as XRF, XRD, TGA, slagging indices, quantitative AFT tests and phase diagrams. The results from this work are helpful for understanding the size dependence of ash thermal behavior, and predicting the ash fusion propensity in future practical application of superfine pulverized coal.

2. Experimental section

2.1. Materials

Two bituminous coals from different Chinese coalfields, Shenhua (SH) and Neimongol (NMG), were selected for the investigation. The properties of the raw coals have been discussed extensively in our previous works [23–27], which were comminuted into different equivalent mean particle sizes of 14.7, 17.4, 21.3 and 44.2 μm for SH samples, while 12.5, 14.9, 25.8 and

52.7 μm for NMG coals. The standardized ash preparation procedure (GB/T212-2008, China) was adopted here. Approximately 1 g of air-dried coal particles were placed in a muffle furnace, which was raised to 850 °C slowly and kept for 2 h. Then the ash was collected and sealed in a vessel for analysis.

2.2. Apparatus and procedure

2.2.1. Chemical analysis methods

XRF-1800 (Japan) was utilized to analyze the chemical composition of coal ash, equipped with the Be window and Rh radiation source. Both major and trace elements were discovered in coal ash, which were reported on the basis of oxides such as SiO_2 , Al_2O_3 , MoO_3 , and SrO . These trace elements are associated with inorganic matter, which are concentrated in the combustion residues [28]. Therefore, the interpretation of ash thermal behavior is of great importance to the coal utilization.

The mineralogy analysis of coal ash was performed on a D/max2200 powder diffractometer (Japan), equipped with the radiation source of $\text{Cu K}\alpha$ (1.5406 Å). All the data were collected at the ambient atmosphere, with the 2θ range of 10–70°. The scanning speed was set at 4°/min, with a step size of 0.02°. The morphology of mineral matter in coal was determined from XRD curves using MDI Jade 5.0 software package.

2.2.2. Simultaneous thermal analysis methods

Thermogravimetric analysis (TGA) is a widely accepted method in characterizing the thermal behavior of ash materials [10,29,30]. The evaluation of thermal behavior of coal ash was carried out on a Q600 simultaneous differential scanning calorimeter (DSC)-TGA (America) instrument. All the TGA tests were performed on the coal ash samples that were heated at a constant heating rate of 20 °C/min from the ambient temperature to 1000 °C. An oxidizing atmosphere (i.e., ambient environment) was adopted to be consistent with the AFT tests.

2.2.3. AFT analysis methods

The ash fusion processes were recorded by a Leitz high-temperature microscope (German), equipped with a high-definition camera. The ash samples were moulded into a cube shape (ca., 3 mm \times 3 mm \times 3 mm), which were heated up to 1500 °C in air, with a heating rate of 10 °C/min. Four characteristic temperatures are commonly recognized through visual observation according to the distinct deforming stages of the ash pellets. To improve the insufficiency of the conventional AFT method, an image processing platform was constructed to extract the numerical information such as geometrical property from AFT results. Thus, a complete deformation profile of the whole ash fusion process could be created with high precision and reproducibility.

The main procedures of the image analysis method include the binarization transformation by Photoshop software, the segmentation calculation implemented through Matlab package, and the geometrical information extraction utilizing Image-Pro program. The detailed description can be found in our previous work [31]. Then, the geometrical parameters such as the area, diameter, perimeter, width, length, and fractal dimension, were obtained from the images of the ash pellet recorded during the fusion process. In this paper, the mean diameter was chosen to present the profile of the ash, which comprehensively reflected the shape change of the pellets. It is defined as the average length of diameters measured at 2 degree intervals and passing through object's centroid.

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