



## Full Length Article

# Modification of ash fusion behavior of coal with high ash fusion temperature by red mud addition



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## ABSTRACT

The influences of two red mud (RM) samples (ZZ and ZM) on fusion behaviors of three high ash fusion temperature (AFT) coals (FZ, JC and JZ) were investigated. It was evaluated by determining AFTs of coals, RMs, and the mixtures of coal ash with RM, as well as their compositions of ashes and mineral. The results showed that the contents of CaO, Fe<sub>2</sub>O<sub>3</sub>, and Na<sub>2</sub>O in RMs were higher than those in coal ashes, which caused RMs' AFTs lower than those of coals. The formation of high melting-point mullite might be the main factor causing coal AFTs higher. With the increase in RM mass ratio, the increasing amount of low-melting eutectic (anorthite (CaAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>), fayalite (Fe<sub>2</sub>SiO<sub>4</sub>), and anorthite (Na-rich) (Ca,Na)(Si,Al)<sub>4</sub>O<sub>8</sub>) and amorphous matter made AFTs lower. ZZ reduced AFTs obviously than ZM because the content of Na<sub>2</sub>O in ZZ was higher than that in ZM. Owing to lower ion potential of Na<sup>+</sup> than those of Ca<sup>2+</sup> and Fe<sup>2+</sup>, Na<sup>+</sup> is easier to enter into the lattice of mullite and results in the reduction of AFTs.

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## 1. Introduction

Coal, approximately accounting for 80% global total fossil fuels, is the most important resource for power generation worldwide [1]. Coal directly combustion for power generation has led to series of problems (the global warming and environmental pollution). In order to solve these problems, the power plants combined with Integrated Coal Gasification Combined Cycle (IGCC) with the characteristics of coal clean utilization and wide flexibility in feedstock (coal, biomass, garbage, etc.) has become a hot issue worldwide, e.g., Europe in Buggenum (NL) [2] and Puertollano (ES) [3], which both employing entrained-flow gasification.

Entrained-flow gasification is gaining important attentions due to the advantages of wide adaptability to coal and high efficiency [4]. Ash fusion temperature (AFT) is one of the crucial parameters for the design of gasifier and the selecting of its operational conditions. It is generally used to predict the fusibility behavior of coal ash during coal gasification [5]. In a typical entrained-flow gasifier, the optimal flow temperature (FT) of coal is approximately ranges over from 1340 °C to 1400 °C [6], and slag viscosity

is approximately 25 Pa·s between 1300 °C and 1500 °C for its tapping smoothly [7]. However, >57% of coals have high AFT (FT > 1400 °C) in China. These high AFT coals gasify directly in the entrained-flow gasifier might reduce gasification efficiency due to ash accumulation and slag (ash deposition in the radiative section of gasifier [8]) blocking, which make the shutdown of gasification system further [9]. Consequently, it is necessary to reduce coal AFT to meet the requirement of entrained-flow gasifier.

Ash deposition phenomena are influenced by coal type (ash composition, fusion temperature, and the distribution of mineral matter), reaction atmosphere, particle temperature, and so forth [10]. The adjustment of AFT is generally achieved by adding flux [11,12] and coal blending [13]. In the aspect of adding flux, Jing et al. found that the formation of low melting eutectic because of increasing iron, sodium and calcium content resulted in the decrease of AFTs [14]. Vassilev et al. pointed out that the alkaline cation was the donor of oxygen, which could terminate the aggregation of polymer and reduce its viscosity [15]. Akiyama et al. found that MgO played a role in decreasing the molten slag fraction due to MgO involved the formation of solid-phase aluminosilicates [16]. A Na-based fluxing agent Na<sub>2</sub>O and composite fluxing agent (CaO:Fe<sub>2</sub>O<sub>3</sub> = 3:1) were used to decrease the AFTs of Dongshan coal and Xishan coal [17]. In the aspect of coal blending, coal blending experiments of Pingsuo coal and Shenhua coal, Tianci

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coal and Xiaotun coal indicated the variation in mullite and anorthite contents were the main reason leading to the AFT change of mixed coal [18]. Although adding flux and blending coal both can reduce AFTs of coal, they also have disadvantages: the former is easy to make AFT change rapidly; the latter is greatly restricted by high transport cost. Therefore, it is necessary to find cheap complex fluxes to reduce the AFTs of high ash-melting coals.

Red mud (RM) is solid waste from producing aluminum oxide. Every year, nearly 70–120 million tons of RMs are produced worldwide [19]. It is a hazardous material due to its high alkalinity and radioactivity (contain large amounts of alkaline (e.g., iron, calcium, sodium) and small amount of radioactive elements (cesium, strontium, uranium, etc.)) [20,21]. A large amount long-time stack RM not only pollutes the atmospheric environment, also leads to soil alkalization and groundwater contamination, which pose a security risk to the people's safety [22–24]. Therefore, the comprehensive utilization of RM has become a hot spot worldwide. RM has been used in the fields of building materials [25], adsorbent [26], catalyst or catalyst carrier, rare earth metal extraction, and so on [27,28].

In the field of coal conversion, RM was used as a kind of catalyst for liquefaction of coal and biomass [29,30], and hydrogenating anthracene oil [31], as well as a desulfurization agent for sulfur removal from high-temperature gas [32]. However, the application of RM to reduce coal AFT is rarely reported. The aim of this paper was to investigate the effects of RM on coal AFT and its mechanism. It is expected to provide references to adjust the ash fusion characteristics during coal conversion.

## 2. Materials and methods

### 2.1. Raw materials

The three air-dried coals (Fangzheng coal, Jiaozuo coal, and Jincheng coal respectively referred to as FZ, JC and JZ) were provided by Institute of Coal Chemistry (ICC), Chinese Academy of Sciences (CAS). Two kinds of RM on air-dried basis (Zhongzhou and Zhongmei, respectively referred to as ZZ and ZM) were provided by College of Chemistry and Chemical Engineering, Henan Polytechnic University. The five samples were crushed to <0.198 mm, dried at 105 °C for 24 h in a nitrogen (N<sub>2</sub>) atmosphere, and stored in a cabinet dryer before use. Proximate and ultimate analyses determined by Chinese standard (GB/T476-2001 and GB/T212-2012) are shown in Table 1, respectively. All coals had low water content (1.51–3.88%) and the total sulfur content (0.59–1.76%), and high fixed carbon content (53.93–76.76%). Among the three coals, the volatile matter content of FZ was the highest, but the fixed carbon content was the lowest.

**Table 1**  
Proximate and ultimate analysis of coal samples.

Sample	FZ	JC	JZ
<i>Proximate analysis on air-dried basis (wt.%)</i>			
Moisture	3.88	2.35	1.51
Volatile matter	29.89	7.76	8.38
Ash	12.30	15.60	13.35
Fixed carbon	53.93	74.79	76.76
<i>Ultimate analysis on dry ash free basis (wt.%)</i>			
Carbon	78.30	76.73	80.65
Hydrogen	4.14	2.80	2.85
Oxygen	2.40	3.20	1.39
Nitrogen	1.10	0.90	1.17
Sulfur	1.76	0.77	0.59

### 2.2. Ash preparation

Laboratory ashes of coal samples were prepared according to Chinese standard procedures (GB/T1574-2001). The samples were placed in a muffle furnace. The temperature was increased to 500 °C within 30 min, held at 500 °C for another 30 min, then raised to 815 °C, and kept at this temperature for 2 h. Finally, the ashes were removed and cooled to room temperature.

Sample mixtures at different temperatures were prepared as follows. RMs were added into laboratory ashes with different mass ratios of 5, 10, 15, 20, 25, and 30%, respectively. The ash-RM mixtures were mixed until reached uniformity. Mixed ash samples at 1200 °C and the coal ash samples at 815, 900, 1000, 1100, 1200 and 1300 °C were prepared in an ALHR-2 AFT analyzer (Ao-lian Co. Ltd., China) to keep the same condition as their AFTs testing [33]. The samples were taken out at pre-setting temperature, and were quenched immediately by ice water to prevent phase variation and crystal segregation. The quenched samples were placed in a vacuum oven at 105 °C for 36 h, and then were stored in a cabinet dryer before using.

### 2.3. AFT measurements of samples

AFT measurements were conducted on the ALHR-2 AFT analyzer. AFT tests strictly followed Chinese standard test method (GB/219-2008). The ash cone was first heated to 900 °C at 20 °C/min, and then increased at 5 °C/min in reducing atmosphere (1:1, H<sub>2</sub>/CO<sub>2</sub>, volume ratio). During the heating process, the deformation temperature (DT), softening temperature (ST), hemisphere temperature (HT), and FT of ashes were obtained based on the variation of ash cone shape.

### 2.4. Ash analytical methods

Sample compositions conducted on an X-ray fluorescence spectrometer (XRF-1800, Shimadzu, Japan) with a Rh target X-ray tube (50 kV, 40 mA) are shown in Table 2. The mineral composition was performed on an X-ray diffraction (XRD) using a D/max 2400X power diffract meter (Rignaku co., Japan) with the characteristic Cu Ka radiation. Operating conditions were 40 kV, Ka<sub>1</sub> = 0.15408 nm and 100 mA. Scanning speed was 5° 2θ/min with the step size of 0.01° 2θ in the range of 5° and 80°. The mineral compositions were analyzed by the computer software package MDI Jade 5.0.

## 3. Results and discussion

### 3.1. Analysis of the basic properties of five samples

AFTs and the ash compositions of five samples are shown in Table 2. The AFTs of RMs are lower than those of coals. It is clear that the contents of CaO, Fe<sub>2</sub>O<sub>3</sub> and Na<sub>2</sub>O in RMs are obviously higher than those of coal ashes. It was found that the ionic potential of Ca<sup>2+</sup>, Na<sup>+</sup> and Fe<sup>3+</sup> was lower (20.20 nm<sup>-1</sup>, 10.53 nm<sup>-1</sup> and 39.47 nm<sup>-1</sup>, respectively), which prevented polymer formation [34]. They also reacted with other minerals to form low temperature eutectics under reducing atmosphere, thereby decreasing the AFTs [34]. A strong relationship between the AFT and the acid/base (A/B = (SiO<sub>2</sub> + Al<sub>2</sub>O<sub>3</sub> + TiO<sub>2</sub>)/(Fe<sub>2</sub>O<sub>3</sub> + CaO + MgO + Na<sub>2</sub>O + K<sub>2</sub>O + SO<sub>3</sub>)) ratio was found [35]. AFTs increases with increasing A/B ratios [36]. A/B values increase in the order ZZ < ZM < FZ < JC < JZ, which are also shown in Table 2. To a certain extent, this reflects increase order of AFTs of five samples.

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