



Full Length Article

Inhibition of sodium release from Zhundong coal via the addition of mineral additives: A combination of online multi-point LIBS and offline experimental measurements



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ARTICLE INFO

Keywords:

Zhundong coal
Sodium
Alkali metal
Sorbent additives
LIBS

ABSTRACT

The retention performance of 5 different sorbent additives, including 2 pure additives, i.e., silica and alumina, and 3 typical natural mineral additives, i.e., kaolin, mica and pyrophyllite, on the release of sodium during the combustion of Zhundong coal is investigated via a combination of online multi-point Laser-Induced Breakdown Spectroscopy (LIBS) technique and offline measurements including inductively coupled plasma atomic emission spectrometer (ICP-AES), X-ray diffraction, and ash fusion temperatures (AFTs). The online and offline measurement results for the sodium release of Zhundong coal/additives mixtures are compared and verified with each other. Ternary phase diagram simulations are performed to further substantiate the impact of different additives on liquidus temperatures of Zhundong coal ash. All the five sorbent additives show a significant sodium retention effect, while alumina and kaolin are better additives considering the effects on AFTs.

1. Introduction

Coal, as an energy source, supports approximately 40% of the worldwide electricity [1]. Due to its broad availability and the flexibility of coal combustion systems, the utilization of coal is expected to continue in the near and medium future [2,3]. It was known that the presence of sodium in coal can lead to rapid ash deposition on heat transfer surfaces of coal-fired boilers [4]. By reacting with chloride and sulfur species in the gas phase, sodium can also form complex chloride and sulfur compounds, which leads to fouling and corrosion of heat transfer surfaces [5]. These sodium-induced issues significantly restrict the utilization of the low rank coal with a relatively high sodium concentration, e.g., Zhundong coal [6]. The new 390-billion-ton Zhundong coalmine has been recently explored in China. Under the current consumption rate of coal, this new source of coal supply is able to support the energy demand of China for more than 100 years [7]. However, utilization of this new source of Zhundong coal requires reducing the harmful sodium emissions during coal combustion [8].

Better understanding of the release characteristics of sodium plays a key role in the development of these sodium-controlling technologies. In the past decades, research on sodium release during coal combustion mainly employs offline measuring techniques, e.g., sampling

measurements by analyzing the bulk composition of fly ash and ash deposits [9]. According to the pioneering work in the field, the forms of sodium in coal can be categorized to four classes [10,11]: (1) water-soluble sodium, which can be dissolved within moisture and exist as Na salts; (2) NH₄Ac-soluble sodium, which is organically bounded with carboxyl groups; (3) HCl-soluble sodium, which is organically bounded with nitrogen- or oxygen-containing functional groups; (4) insoluble sodium, which is bounded with clay minerals. The first three forms of sodium are releasable during coal combustion while the last insoluble sodium remains in the residual ash due to the low activity of silicate minerals [5]. Although offline measurements can detect the final amount and composition of sodium species at post-combustion stage, it cannot directly capture the dynamic sodium release process and obtain the time-resolved information of the sodium release rate at different coal combustion stages. To overcome these disadvantages of offline techniques, online measurements using advanced laser diagnostics, e.g., Laser-Induced Fragmentation Fluorescence (ELIF) [12], Tunable Diode Laser Absorption Spectroscopy (TDLAS) [13], Laser-Induced Breakdown Spectroscopy (LIBS) [14] and Planar Laser-Induced Fluorescence (PLIF) [15], have been recently implemented. For instance, van Eyk et al. [16] developed a PLIF technique to measure the distribution of atomic Na and its time-resolved release process in the plume of a

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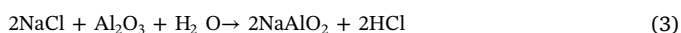
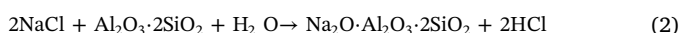
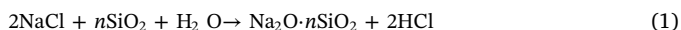
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burning coal particle. LIBS is a type of atomic emission spectroscopy based on the plasma generated by a high power laser. The detailed principle of LIBS can be found in previous publications [17,18]. Recently, simultaneous measurement of sodium and potassium release over burning coal and wood particles using this technique has been reported by Hsu et al. [19] and also in our previous work [14]. However, the abovementioned studies focused on either the measurement of the distribution of one sodium species, e.g., atomic Na, or the measurement of total sodium element at a single measuring point. To our best knowledge, the directly time-resolved measurement of total sodium flux from a burning coal particle via multi-point LIBS technique has not been reported.

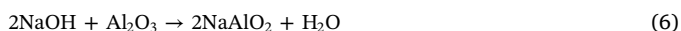
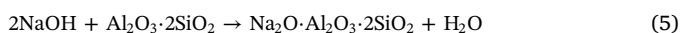
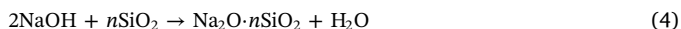
Technologies for reducing the harmful sodium emissions can be implemented prior to, during, or after the combustion process. Washing coal with water, acid solutions, or aluminum salt solutions can reduce the content of sodium compounds prior to combustion [20,21]. In coal gasification, sorbent filters can be used to clean the product gases and remove sodium compounds [22,23]. On the other hand, pulverized-sorbents can be injected into boilers along with pulverized-coal as additives to reduce sodium release from the coal during combustion. Since this method can be directly applied in the current combustion systems without any major reconfiguration, it has attracted a lot of attentions [24–32]. Previous studies on additives for the retention of sodium release found that silica (SiO_2) and alumina (Al_2O_3) are the two important and effective compounds. Lee and Johnson [24] investigated sodium retention in pressurized fluidized bed gasification (PFBG), and their study showed that activated bauxite (81.5% Al_2O_3 , 10.0% SiO_2) can retain 98% of volatilized NaCl. Punjak and Shadman [25] studied the effects of kaolin and bauxite on sodium retention, and they found that the retention by kaolin is irreversible. Takuwa and Naruse [26] further investigated the sodium retention by kaolin via a drop tube furnace. It was suggested that NaOH is the major sodium species in the coal used and can be reduced by reacting with Al_2O_3 in kaolin. Kosminski et al. [29] noted that molten sodium carbonate is the dominant sodium species between 800 °C and 850 °C in the low-rank coal and silica can react with it to reduce sodium release.

Kyi and Chadwick [27] evaluated the performance of various additives, e.g., silica (diatomite), alumina, calcium silicate (wollastonite), mine overburden, and various aluminosilicates with different alumina/silica ratios on the sodium retention of different coals. From their results and previous studies [24,26], the reactions of sodium retention can be summarized as follows:

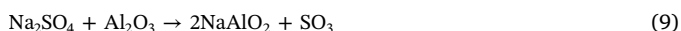
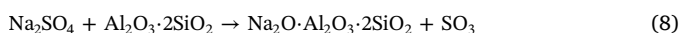
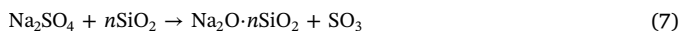
For NaCl:



For NaOH:



For Na_2SO_4 :



The abovementioned previous studies on additives for sodium retention employed offline measurements to monitor the amount of sodium released, and therefore there is no information about the temporal release characteristics of sodium for coal blended with additives. Moreover, the sodium retention performance of additives at different stages of coal combustion has rarely been revealed yet.

In our previous work [33], single point LIBS measurements were performed to obtain the gaseous sodium concentration at one fixed position during the combustion of Zhundong coal blended with different additives. In the present study, the performance of different sorbent additives on the release of sodium during the combustion of Zhundong coal will be investigated by combined online- and offline-measurements. Online multi-point LIBS technique, upgraded from the single point LIBS, will be employed for the in-situ measurement of the time-resolved sodium release flux of the coal and coal/additives mixtures. Offline techniques, which include inductively coupled plasma atomic emission spectrometer (ICP-AES), X-ray diffraction, and ash fusion temperatures (AFTs), will be applied to investigate the individual behavior of each sodium class and the influence of additives on the compositions and slagging characteristics of residual ash. The offline ICP-AES results will serve as a reference to verify the online quantitative multi-point LIBS measurements. Based on these experiments, the advantage and disadvantages of different additives for sodium retention of Zhundong coal have been discussed.

The paper is organized as follows: details on the online- and offline-measurement methods are presented in Section 2. Global sodium retention characteristics of different additives revealed from the offline ICP-AES measurements are illustrated in Section 3.1. The temporal sodium release characteristics of different blended coal/additive pellets obtained using the online multi-point LIBS technique are discussed in Section 3.2. The subsequent Section 3.3 is devoted to analyze the effects of additives on the chemical compositions and AFTs of residual ash. Finally, the findings of this paper are summarized in Section 4.

2. Experimental

2.1. Coal and mineral additives

A typical Chinese brown coal called Zhundong coal [5,8,33–36], which has appreciable sodium content, is considered in the present study. The results of proximate analysis and ultimate analysis, as well as the ash composition of the coal sample are listed in Table 1. Proximate analyses were performed by chemical analysis methods according to the Chinese National Standard GB/T 212-2008. Ultimate analyses were determined according to the Chinese National Standard GB/T 476-2008 (for C, H), GB/T 19227-2008 (for N) and GB/T 214-2007 (for S). The mass fraction of oxygen (O) was determined by 100%-C%-H%-N%-S%. Ash compositions were determined according to the Chinese National Standard GB/T 1574-2007. Different additives were blended with Zhundong coal to explore their effects on sodium retention. Specifically, typical natural mineral additives, i.e., kaolin, mica and pyrophyllite, and pure silica and alumina are employed. The physical and chemical properties of these additives are summarized in Table 2. All the additives are blended with the coal at a dosing ratio of 3% by weight, which is a typical dose for sorbents [33]. The mixing process of coal and additives is as follows. Coal and additives are first grounded and sieved to a fine powder (< 75 μm). Then they are mixed according to the dosing ratio in a grinder of 32,000 rpm for 5 min to produce coal-additive mixture powder.

Table 1
Chemical analysis of Zhundong coal.

Proximate analysis (wt%, air dry basis)						
Moisture	Ash	Volatile	Fix Carbon			
9.85	4.23	28.72	57.2			
Ultimate analysis (wt%, dry ash free basis)						
Carbon	Hydrogen	Nitrogen	Sulfur	Oxygen		
79.29	2.89	0.88	0.43	16.5		
Ash composition (wt%)						
SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O
10.80	9.62	3.95	36.82	9.20	0.40	10.87

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