

Experimental Investigation of Gaseous Sodium Release in Slag-Tapping Coal-Fired Furnaces by Spontaneous Emission Spectroscopy

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Abstract: High-alkali coal is rich in alkali metals, which can cause serious effects such as slagging and corrosion on the heating surface during combustion and utilization. A portable spectral system was utilized to simultaneously measure gaseous Na concentration and temperature in a 20 kW slag-tapping combustor and a slagging boiler furnace of a 300 MW power generation unit by flame spontaneous emission spectroscopy (FES) for simultaneous measuring. The result shows that both ZD-FK and ZD-HSQ (Fukang coal and Hongshaquan coal, Xinjiang Zhundong high-alkali coal) combustion flame temperatures are around 1400 °C at the outlet of the cyclone burner while the latter is slightly higher. The sodium concentration in the gas phase increases with the rising of the initial combustion temperature and unit load for one kind of coal, and the level of sodium concentration has a strong correlation with the Na content for different coal. Most of the sodium in the high temperature zone of the furnace exists in the form of gas phase, and more sodium migrates to fly ash. Combined with the analysis of fly ash and liquid slag samples, a closed-loop analysis of the Na migration path could be established.

Keywords: slag-tapping furnace; high alkali coal; gaseous sodium concentration; temperature; spontaneous emission spectroscopy



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1. Introduction

The gaseous alkali metals, mainly including sodium (Na) and potassium (K) released from the combustion of high alkali fuels such as Zhundong coal and biomass, causes fouling and slagging problems on heating surface of furnaces, which can significantly affect the operation safety [1,2]. A past investigation found that much sodium could be retained with the molten slag in the fuel-rich primary combustion zone in a slagging combustor [3]. In a recent research, the slagging-type cyclone combustion is also proved to be suitable for the Zhundong coal combustion because of the suppression of the molten slag on sodium diffusion [4]. A following experimental investigation in a slag-tapping combustor studied the sodium retention behavior of Zhundong high-alkali coal, which also shows that the molten slag has a high capacity of capturing alkali metals released during the slag-tapping combustion [5,6]. The release of vapor phase sodium has strong dependence on combustion temperature and coal composition in the slag-tapping combustion. Consequently, quantitatively determining the concentration of gaseous alkali metals and temperature in combustion process is of great significance for increased understanding of the alkali release.

Recent activities in numerical simulation and experimental research on thermochemical transformation of biomass were reviewed by Fatehi et al. [7], in which state-of-the-art optical diagnostic methods have been developed or applied to detect temperature and key species (alkali metals, etc.) involved in biomass thermochemical conversion. Measurement techniques widely used in monitoring the release of alkali species (mainly K and Na), can be divided into active type and passive type. The former is a laser-based technique mainly including Laser-Induced Breakdown Spectroscopy (LIBS), Tunable Diode Laser Absorption Spectroscopy (TDLAS), Planar Laser-Induced Fluorescence (PLIF), and Colinear Photo-Fragmentation and Atomic Absorption Spectroscopy (CPFAAS), while the latter is based on the analysis of flame spontaneous emission spectroscopy (FES). Observation of spontaneous emission during the combustion of highly alkali fuels in high-temperature environments is a non-invasive, easy-to-implement, and low-cost method to investigate the release of alkali metals [8]. FES was employed for the first time to observe the potassium release of biomass particles combustion [9]. Following the previous research, Mason et al. [10,11] obtained the potassium temporal release patterns for 13 different solid biomass fuels and developed a K-release model of single biomass particle combustion. Lim et al. [12] found that the Na/K ratio from the atomic emission spectra can be used as a tentative in situ indicator of the respective atoms' content. In addition, FES can also be applied to calculate flame temperature through analyzing the multi-wavelength emission spectrum in the visible and near-infrared region [13,14].

Based on the theory of FES, He et al. [15] simultaneously measured the concentration of gaseous phase potassium, temperature, and thermal radiation during the whole process of biomass pellets combustion by means of the analysis of spontaneous emission spectra and a calibration procedure. Lou et al. [16,17] developed a portable spectral system and extended the application of FES on in situ measurement of alkali metals release in municipal solid waste incinerators and coal-fired boiler furnaces. Li et al. [18,19] improved the calibration procedure of FES and established a functional model between alkali metals concentration, temperature, and spectral radiative intensity of alkali metals. Besides that, Paulauskas et al. [20] studied the K and Na release behavior during biomass combustion by using FES and found the release profiles by the FES method to correlate with the data from the ICP-MS. Li et al. [21] predicted potassium concentration in biomass fuels by using neural network analyzing spectroscopic features of flame and potassium.

The above research achievements on the measurement of alkali metals in the combustion process inspired us to employ the FES approach for investigation of gaseous sodium release in slag-tapping coal-fired furnaces. Therefore, a portable spectral system similar to that in Refs. [15,16] was utilized to simultaneously measure gaseous Na concentration and temperature in a 20kW slag-tapping combustor and a slagging boiler furnace of a 300 MW power generation unit. The effects of combustion temperature and coal composition on the release of sodium were analyzed. In addition, combined with X-ray fluorescence (XRF) analysis, some simple conclusions about the migration route of sodium were drawn.

2. Method and Experimental Setup

The principle of FES for simultaneously measuring gaseous alkali metals concentration and temperature has been introduced in detail in Refs. [15–19]. The spontaneous emission spectra during high alkali fuels combustion can be collected by emission spectrometers. Through a calibration procedure in blackbody furnaces [13], the raw outputs (count values) of the spectrometer, which represent the relative value of flames spectral radiation intensity, are transferred to absolute spectral radiative intensities along the wavelength. The obtained spectral radiative intensities of high alkali fuels combustion flame consist of continuous spectra by thermal radiation and discontinuous spectra due to the emission line of gas phase alkali metals [22]. The combustion temperature can be calculated by the method of multi-wavelength radiation analysis of the continuous spectrum in the visible and near-infrared wavelength ranges [8,13,14].

The characteristic line intensities spectra of alkali metals (Na and K) in flames can be calculated by deducting thermal radiation from the collected flame spectra. Then, according to flame emission spectroscopy [23,24], Na and K released into the gas phase in the flame are thermally excited, so the spectral intensities of the emission lines are considered to be proportional to the concentrations of volatilized Na and K in the flame, respectively. Through another calibration experiment, implemented in a Hencken flat flame burner, a function relationship between gaseous Na or K concentration, temperature, and spectral emission intensity can be established [18,19]. Thus, with known spectral intensities of the emission lines of alkali metals and temperature calculated by multi-wavelength radiation analysis, the gaseous Na or K concentration could be obtained.

A portable spectral system containing a visible spectrometer (Type: AvaSpec) assembled with a fiber, a collimating lens, and a Tablet Personal Computer (TPC) has been applied to simultaneously measure the gaseous alkali metals concentration and combustion temperature in furnaces. The measurement wavelength range of the spectrometer is from 360 to 1100 nm with the spectral resolution of 0.5–0.7 nm. As shown in Figure 1, the portable spectral system was placed on a tripod near a viewing port of the furnace in the experiment. The flame emission spectra were collected by the spectrometer, then converted into digital signals, and finally sent to the TPC via a USB cable. A dedicated application software developed by the authors was installed in the TPC to process the flame spectrum and determine the temperature and gas phase alkali metal concentration.

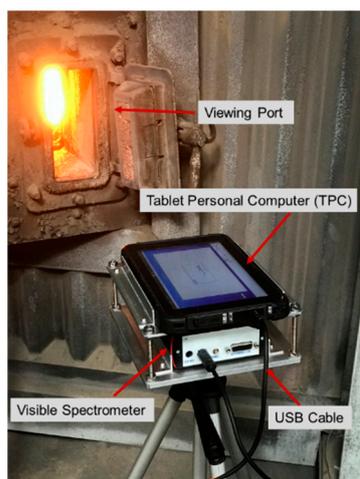


Figure 1. Photo of the portable spectral system placed on a tripod near a viewing port of a furnace.

3. Experiment in Slag-Tapping Combustor

3.1. Experimental Conditions

The experiment was firstly conducted in a 20-kW slagging-type cyclone fired combustor test facility, which was introduced in Refs. [5,6]. As shown in Figure 2, the test facility is mainly composed of primary air inlet, secondary air inlet, electrical heating cyclone chamber, burn-out chamber with a partition panel inside and a slag tapping hole at the bottom, and other auxiliary equipment. The pulverized coal with primary air was injected into the cyclone chamber via a swirl burner. The secondary air was injected from the secondary air inlet on the top of cyclone chamber and caused a strong swirling flame upon mixing with coal particles. To study the influence of the combustion temperature on sodium release from coal, the combustion temperature could be adjusted through the electric heater to a specific temperature between 1100 °C and 1500 °C with an interval of 100 °C. The larger coal particles burned during the cyclone-fired process, while some fine coal or ash particles entered into the burnout chamber with hot flue gas. The liquid slag flowed out through the tapping hole. There is a viewing port on the side wall of the burn-out chamber, through which the flame spectra were obtained by the portable spectral system.

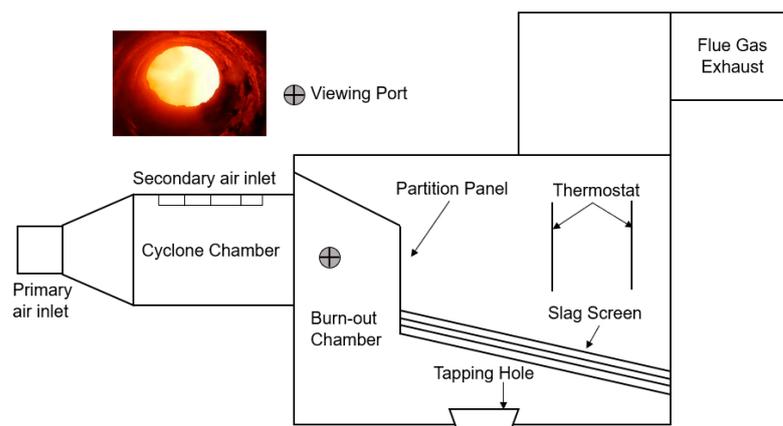


Figure 2. Schematic diagram of the slagging-type cyclone fired combustor test facility.

Two types of Zhundong coals, labeled as ZD-FK and ZD-HSQ, were used in the experiment. The properties of coals are analyzed in Table 1. Both coals are low ash ignites. The Na content in the ash of ZD-HSQ is higher than that of ZD-FK. The test conditions are listed in Table 2. To investigate the effect of combustion temperature on sodium release, the two coals were burned at two different kinds of temperature, respectively.

Table 1. The properties of coals used in the slag-tapping combustor.

Coal	Proximate Analyses (Air Dried Basis, wt%)				Ultimate Analyses (Air Dried Basis, wt%)				
	Moisture	Volatility	Ash	Fixed Carbon	C	H	O	N	S
ZD-FK	16.72	23.77	5.59	53.92	74.42	1.96	15.69	0.62	0.49
ZD-HSQ	14.63	32.82	7.52	45.03	61.52	3.59	11.48	0.84	0.42
Ash Composition (wt%)									
Coal	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	K ₂ O	Na ₂ O	SO ₃	TiO ₂
ZD-FK	10.37	5.27	12.36	35.59	10.01	0.39	1.90	19.9	0.72
ZD-HSQ	31.52	13.32	18.68	16.63	6.19	0.69	3.18	7.66	0.68

Table 2. Test conditions in the slag-tapping combustor.

Case	1.1	1.2	1.3	1.4
Coal	ZD-FK	ZD-FK	ZD-HSQ	ZD-HSQ
Combustion temperature	1100 °C	1300 °C	1200 °C	1400 °C

3.2. Results and Analysis

According to the test conditions, shown in Table 2, when the load is stable, the portable spectral system is used to collect data at the viewing port for more than 300 s. And the average value of the test results within the measured times are taken as the final results, as shown in Figure 3. The standard deviation is indicated by the flame fluctuation during the measurement time.

When high alkali coal is burned, most of the organic sodium will be released in the gaseous phase. As can be seen from Figure 3, under the two cases of burning ZD-FK coal condition, the average temperature at the viewing port along the line-of-sight are 1386 °C (Case 1.1) and 1420 °C (Case 1.2), respectively, while it is 1398 °C (Case 1.3) and 1436 °C (Case 1.4) for the ZD-HSQ coal. For the same kind of coal, there are certain differences for the measured temperature. When the combustion temperature is lower, the gas-phase sodium concentration also decreases, indicating the strong dependence of temperature on the gaseous phase sodium release. Despite the difference in initial heating combustion temperature, there was no significant change in the flame temperature for ZD-FK and

ZD-HSQ coals, but the gaseous phase sodium released for ZD-HSQ coal is significantly higher than that of ZD-FK coal, even under the condition where the initial combustion temperature was lower (Case 1.2) than that of ZD-FK coal (Case 1.3).

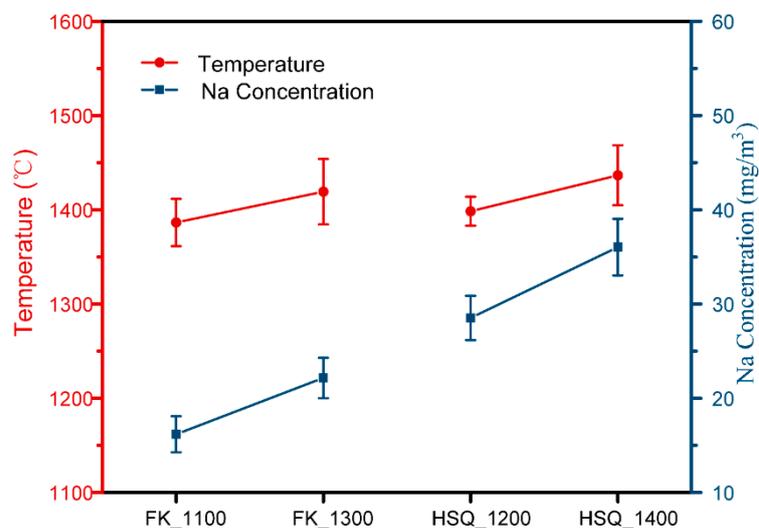


Figure 3. Measured gaseous Na concentration and temperature with standard deviations.

Almost all sulfur-containing compounds and some sodium-containing compounds remain gaseous at temperatures above 1200 °C [25]. From the temperature results, it can be seen that under different test conditions, the flame temperatures of FK and SH combustion both exceed 1200 °C. Therefore, except for a part of Na in the form of aluminosilicate, the rest of the gas-phase Na released in the form of elemental, oxidized, and chlorinated states is still in the gaseous stage. According to Table 2, the sodium content of SH coal is higher than that of FK coal, the content of organic sodium in coal directly affects the concentration of gaseous Na in flue gas and the combustion temperature of SH coal is higher, which promotes the release of Na in the gaseous phase. Despite the ZD-HSQ has higher Si and Al content, part of Na reacts with silica to form insoluble sodium-based silicate, and the formation of sodium silicates reduces the amount of gaseous Na, but from the test results, it can be considered that this proportion is small.

In order to further investigate the relationship between gas phase sodium release and temperature, the results during the measuring time under each working condition were analyzed. Generally, the stability of combustion in the furnace refers to relative stability. The measured results obtained by the portable spectral system are variable and fluctuate up and down a certain result, which also reflects the instability of the flame. The results were arranged in ascending order of temperature, and the variation trend of Na concentration with temperature was obtained. The gas phase Na concentration were smoothed by filtering, as shown in Figure 4. It can be seen that the sodium release increases with the rising of the combustion temperature under the four working conditions, showing a positive correlation trend. Further, Figure 5 shows the results of the smoothed Na concentration changes with temperature. At the same flame temperature, the gas Na concentration is different, and perhaps the different initial temperature affects the burnout degree of coal. In addition, when the temperature reaches a certain value, the increasing trend of Na concentration becomes gentle, which indicates that the gas phase Na release reaches the extreme value at this temperature.

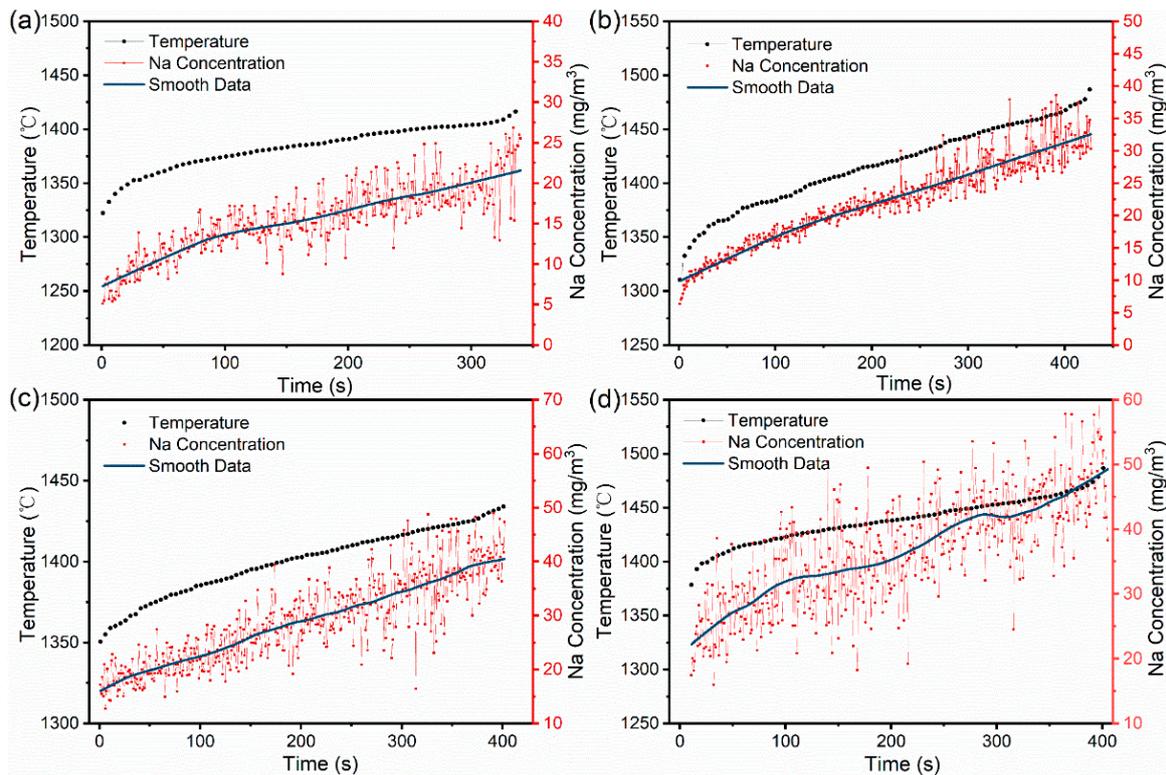


Figure 4. Changes in gaseous phase Na concentration and temperature during measurement time (in ascending order of temperature), (a) FK_1100, (b) FK_1300, (c) HSQ_1200, (d) HSQ_1400.

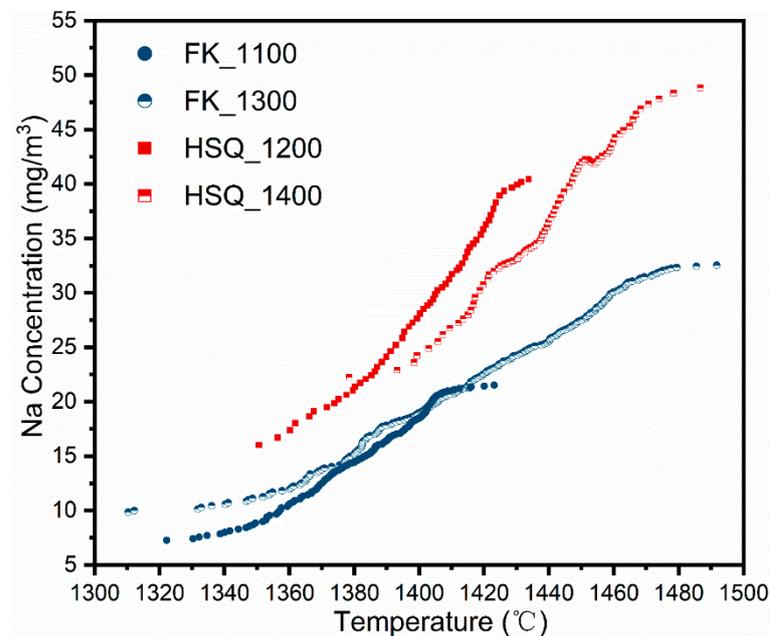


Figure 5. Variation of Na concentration with temperature during the combustion of different types of coal.

4. Experiment in Slagging Boiler Furnace

4.1. Experimental Conditions

Measurement of gaseous Na concentration and temperature in a cyclone coal-fired boiler with slag-tapping furnace of a 300 MW power generation unit was also conducted by using the portable spectral system. The cyclone boiler is an ultra-high pressure, once-

through boiler with a rated capacity of 830 t/h that is made by the BABCOCK company. As shown schematically in Figure 6, two burnout chambers, a slag chamber, and a vertical flue are symmetrically arranged on both sides of the boiler to form a double U-shaped furnace tower structure. Eight cyclone burners are staggered on top of each combustion chamber. Each cyclone burner is equipped with one primary air inlet and two secondary air inlets, and coal particles are injected together with the primary air. Each coal mill, named #10, #20, #30, and #40, controls four burners. The viewing ports labeled as #1 and #2 at the same location in both burnout chambers are used as the measuring points.

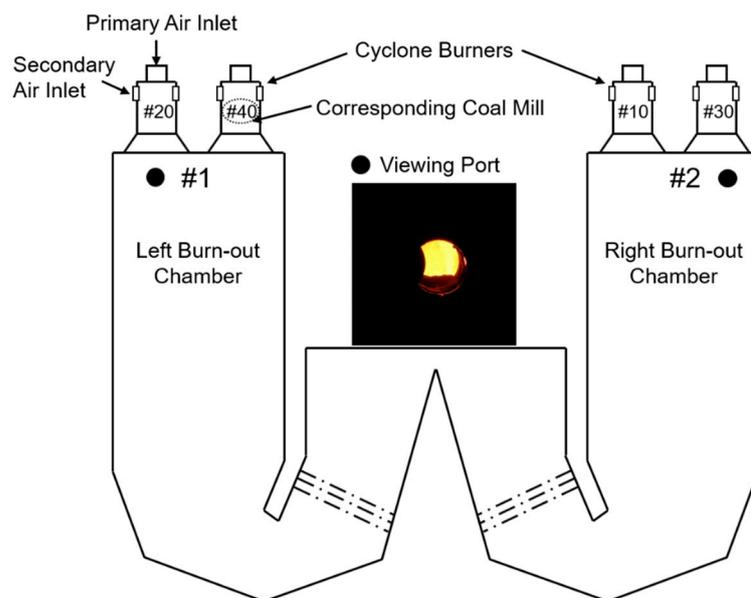


Figure 6. Schematic diagram of the boiler.

The coal tested in this experiment is Shenhua coal (SH). The proximate analysis, ultimate analysis, and ash composition analysis of SH are shown in Table 3. The content of Na_2O in the ash is 2.0%. Experiments were carried out under the conditions of medium and low load of the raw coal and raw coal with adding salt, that is, a certain quality of soda ash was added at the inlet of the coal feeder, and then mixed with the raw coal in the coal mill evenly and sent to the cyclone burner through the primary air to simulate the combustion of high alkali coal. The mass of added soda ash is determined according to the converted Na_2O mass fraction in low temperature ash of 3.5% and 5.0%, respectively. The specific test conditions are shown in Table 4. In the table, coal mill on indicates which coal mill is running.

Table 3. The properties of coals used in the slag-tapping combustor.

Coal	Proximate Analyses (Air Dried Basis, wt%)				Ultimate Analyses (Air Dried Basis, wt%)				
	Moisture	Volatility	Ash	Fixed Carbon	C	H	O	N	S
SH	2.21	29.86	13.42	54.51	68.62	4.19	10.29	0.83	0.44
Ash Composition (wt%)									
Coal	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	K_2O	Na_2O	SO_3	TiO_2
SH	57.67	21.87	5.17	5.76	1.46	2.23	2.00	0.16	0.87

Table 4. Operating conditions of the test.

Case	Coal Mill On	Load (MW)
SH	#20, #40, #10	240
SH+3.5%Na ₂ CO ₃	#20, #40, #10	251
SH+5%Na ₂ CO ₃	#20, #40, #10	224

4.2. Results and Analysis

Under the conditions of the unit load stable, the furnace flame was measured at the two viewing ports for 600 s. The data collection frequency is once per second. The average value of the results during the measuring time was taken as the final result.

Figure 7 shows the measured results of two viewing ports under medium load conditions. The interpretation of standard deviation is consistent with the previous description. The average temperature of #1 is about 60K higher than that of #2. This is mainly due to only one coal mill (#10) working for the burn chamber where #2 is located. In addition, there is no significant change with the increase of salt content. Obviously, there is a large difference in gaseous phase sodium concentration at two measuring points. Under the condition of burning raw coal, the gaseous phase sodium concentration is 37.9 mg/m³ and 6.9 mg/m³, respectively. This is mainly caused by the difference in the group of coal mill working in the burning chamber where the two viewing ports are located. On the one hand, the total mass of coal entering the furnace increases, and, on the other hand, the temperature rising promotes the release of sodium during coal combustion. Otherwise, as the proportion of added salt increases, the gaseous phase sodium concentration increases too. The standard deviation of #1 is significantly higher than #2, which is also mainly related to the coal feeding of each chamber.

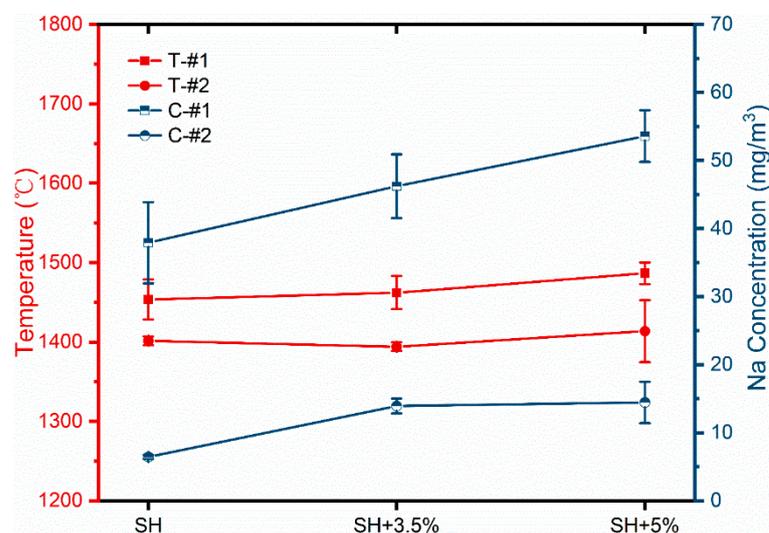


Figure 7. Measured gaseous Na concentration and temperature with standard deviations of the two viewing ports under middle unit load condition.

The ash produced by inorganic matter during coal combustion is the main source of ash deposition and liquid slag. At the same time of testing, liquid slag and fly ash were collected at the bottom of the left burn-out chamber and out of the flue of the furnace under middle unit load condition, respectively. The composition of fly ash and crushed liquid slag were analyzed by X-ray fluorescence (XRF).

Figure 8 shows the element distribution of liquid slag and fly ash in the form of oxides. The major oxides in the liquid slag and fly ash of SH coal are SiO₂, Al₂O₃, CaO, and Fe₂O₃, while the higher content of Al₂O₃, SO₃ and Na₂O are found in fly ash. This illustrates that under the extremely high combustion temperature in the burn-out chamber,

all sulfur-containing compounds have been decomposed almost completely and exist in gaseous form in the flue gas. The relative proportions of Fe_2O_3 and SiO_2 in the fly ash decrease, while the Na content increases, indicating that in this gas temperature range, silicon and aluminum oxides have a high capture effect on alkali metals. At the same time, due to the combination of gaseous phase sodium components and SO_2 in the flue gas, it begins to condense and precipitate from the flue gas at a temperature of 1000°C , adhering to the surface of ash particles or agglomerate and deposit, resulting in the mass percentage of S element in the fly ash sample rise.

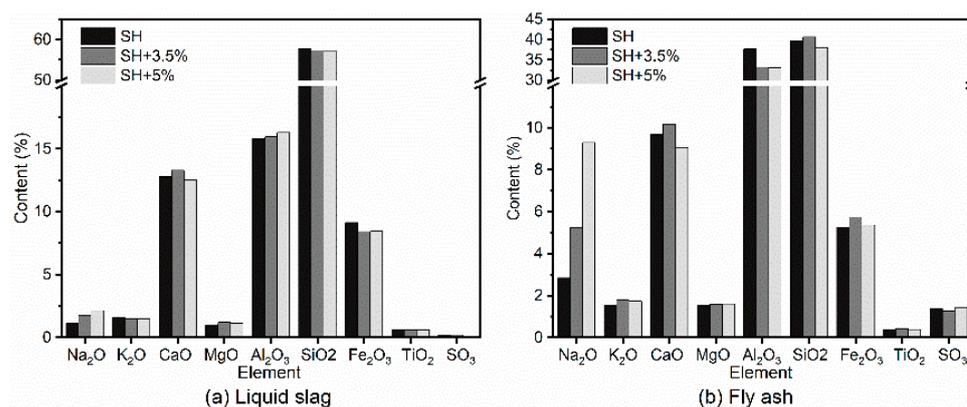


Figure 8. Element distribution of liquid slag and fly ash.

Moreover, with the increasing sodium salt addition, the Na content in both liquid slag and fly ash increases, but the latter is much higher, and the content of other elements does not change significantly. Obviously, the temperature of the liquid slag formation area is significantly higher than that of the flue where the fly ash is located. The sodium in the liquid slag formation zone is more in the gas phase, so that the sodium content is less. At the same time, due to the high capture rate of slag in the cyclone furnace, most of the inorganic sodium is volatilized into the flue gas, which is the same as previously described in the gaseous phase sodium measured results, thereby increasing the relative content of alkali metals in the fly ash.

From the above analysis, it can be seen that the higher proportion of sodium in coal, the more gaseous phase sodium is released during combustion, but it mainly migrates to fly ash. In addition, almost all the sulfates in coal are decomposed, the liquid slag is enriched with Si and Fe, and most of the sodium is volatilized into the flue gas.

5. Conclusions

In this paper, a portable spectral system was used to measure the temperature and Na concentration in the outlet area of the cyclone burner of a 20 kW slag-tapping combustor and a slagging boiler furnace of a 300 MW power generation unit, and the influence of temperature and coal composition on sodium release was reported. The main conclusions are as follows:

- (1) The gas-phase Na concentration released from combustion different kinds of coal, and has a strong correlation with the combustion temperature and the content of organic Na in the coal. Higher concentrations of gaseous sodium were still present in the cyclone burner outlet area. The gaseous phase Na concentration can reflect the change of load. Under a higher unit load, the flame temperature and gaseous phase Na concentration increase significantly at the outlet area cyclone burner.
- (2) The Na content in liquid slag is lower than that in fly ash. The gas phase Na released from the coal is more condensed in the low temperature region of the flue gas and migrates into the fly ash. Si and Fe are more enriched in liquid slag, while S is almost in fly ash;

- (3) Through the measurement of flame temperature and gas Na concentration at the outlet of the cyclone of the liquid slag discharge furnace by FES approach, combined with the analysis of fly ash and liquid slag samples, a closed-loop analysis of the Na migration path can be established.

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