



Release of alkali metals during co-firing biomass and coal



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ABSTRACT

The release of alkali metals is investigated by means of ash element measurement and chemical equilibrium calculation during co-firing biomass (cornstalk derived fuel) and coal. The experiments are operated under different mass blending fraction and temperature conditions. With increasing cornstalk fraction, the normalized ash content decreases according to a quadratic curve because the formation of alkali aluminosilicate may result in more elements being retained in ash. Similarly, the release ratios of K, Na, and S do not change linearly with the variation of cornstalk fraction either, which are suppressed significantly by the interaction of biomass and coal ashes. For the effect of CaO additive, the release ratios of K and Na increase with its enhancement. The equilibrium analysis is used to predict and evaluate the release of alkali metals. The release of alkali metals enhances with the increase of temperature and cornstalk fraction. For silicon-lean blending fuels, adding Ca will decrease HCl(g) and release more KCl(g) and KOH(g) in both fuel-rich and air-rich conditions. More KCl(g) and NaCl(g) will be formed with increasing Ca/(S + 0.5Cl) ratio due to the formation of CaSiO₃(s) and the reduction of alkali aluminosilicate.

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1. Introduction

In recent years, co-firing of biomass and coal has been a hot spot in the field of electricity generation [1–6]. The thermal utilization of biomass, as a substitute or supplement to conventional fossil fuels, not only decreases CO₂ emission in power plants, but also reduces a certain amount of NO_x and SO_x emissions due to the lower content of N and S in biomass [7–9].

Biomass contains high quantities of K, Na and Cl compared with coal. During burning large quantities of biomass, alkali chlorides are released through vaporization. And the gaseous species may subsequently participate in chemical reactions or physical transformations, condensing to form fly ash or aerosols, and finally deposit on heat transfer surfaces [10–14]. Due to the interaction of chlorine and alkali metals in the ash and gas, the complex metal reactions will occur and results in severe corrosion on heat transfer surfaces [10]. Some experimental data have been reported on the ash deposition during biomass combustion [15–17]. Generally, the deposition is caused by condensation, impact of large particles

(>10 μm), and thermophoresis of small particles (<10 μm). The lower melting point for some K species (e.g. KCl melts at 1044 K) implies a high risk of deposition on furnace walls and convection tubes. Thus, fuel ash has been investigated to prevent the release of gaseous alkali-metal compounds [18–21].

The release of gaseous alkali metals during combustion can be measured by direct sampling, surface ionization detector, molecular beam mass spectrometer (MBMS), on-line excimer laser induced fragmentation fluorescence (ELIF), laser-induced breakdown spectroscopy (LIBS), and in-situ alkali chloride monitor (IACM), etc. [22–26]. Monkhouse [22] reviewed on-line systems for detection of metals, particularly alkali metals, such as K and Na in combustion and pyrolysis gases. Porbatzki et al. [23] used MBMS to measure the release of alkali metals, chlorine and sulfur of four different types of biomass, and they found that the release of inorganic species like HCl, KCl and H₂S are strongly dependent on other inorganic constituents in the samples (e.g. Si and Ca). Erbel et al. [24] described the first on-line, in-situ measurement of alkali species in biomass gasification using ELIF. Fatehi et al. [25] studied on a joint numerical and experimental investigation of the release of K from biomass during gasification process. In their work, LIBS was adopted to measure the concentration of K in biomass. Forsberg et al. [26] developed IACM for measurement of alkali chlorides

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in hot flue gas, and they also described the principle and calibration of IACM.

Yang et al. [27] investigated the effect of coal sulfur on the behavior of alkali metals during co-firing biomass and coal. The results show that FeS_2 addition significantly enhances the formation of potassium sulfate when S/K molar ratio is less than two. Meanwhile, increasing FeS_2 dosage reduces the formation of KCl(g) and KOH(g) , and increases the release of HCl(g) . Novakovic et al. [28] studied the release of potassium in K–Ca–Si and K–Ca–P systems, respectively. The results indicated that the presence of water in the gas flow enhances the K-release rate significantly. Rahim et al. [29] investigated the Cl release during slow pyrolysis of NaCl-loaded cellulose in the temperature range of 150–400 °C, providing new data to better understand the low-temperature Cl release during biomass pyrolysis. Johansen et al. [30] studied the release of K, Cl and S during combustion of high-chlorine biomass in bench and pilot scale bed. W. Li et al. [31] examined the release behavior of alkali and alkaline-earth metals during oxy-fuel combustion in different gas environments. The results showed that the release rates of Na and K are higher than Ca and Mg in all types of gas environments. R. Li et al. [19] investigated the effects of temperature on the release and transformation of alkali metal species during co-combustion of coal and sulfur-rich wheat straw. The results indicated that the amounts of K and Na during co-combustion can be reduced by Fe, Ti, S, Si and Al in blended fuels.

The aim of this study is to obtain quantitative data on the release of alkali metals during co-firing of biomass and coal using ash element measurement. Biomass and coal are co-fired under different mass blending fraction and temperature conditions. And the effect of CaO additive used for desulfurization on alkali metal release is also investigated. Moreover, chemical equilibrium analysis is used to predict and evaluate the behavior of alkali metals.

2. Methods

2.1. Element and ash analysis

In the experiment, one bituminous coal (Shenmu coal) and one biomass (cornstalk derived fuel) are heated to prepare ash samples.

Table 1 shows the fuel properties of coal and cornstalk derived fuel, which indicates that Cl content in cornstalk is obviously higher than that in coal. However, coal has higher sulfur and ash content than cornstalk. Table 2 shows the ash composition of coal and cornstalk after combustion. K content in cornstalk ash is apparently much higher than that in coal ash, and this may due to the necessity of K for plant growing. Cornstalk ash also has high Si content. The content of Al, S and Ca in coal ash are higher than those in cornstalk ash. In the process of combustion, Al can react with Si to form aluminosilicate, and S can oxidize to form sulfate. Ca can form CaSO_4 when Ca content is high enough, and adding CaO to absorb S is often used reduce the emission of SO_2 . Table 3 shows the content of Ca, K, Na, S, and P in cornstalk, which is analyzed directly by inductively coupled plasma-atomic emission spectroscopy (ICP-AES). In addition, the element content of coal given in Table 3 is calculated by the ash composition in Table 2.

Table 1
Fuel properties.

Fuel	Ultimate analysis (wt%, air-dried basis)							
	C	H	O	N	S	Cl	Ash	Moisture
Coal	67.04	3.69	13.29	1.28	0.44	0.012	9.10	5.21
Cornstalk derived fuel	44.54	5.77	40.15	1.10	0.16	0.338	3.36	3.16

2.2. Experimental apparatus and methods

To investigate the change of ash content during co-firing coal and cornstalk derived fuel, the experiment is operated in a SK2-5-12 medium temperature box-type resistance furnace with power of 5 kW and rated temperature of 1200 °C. A SR1 series PID temperature controller, made by SHIMADEN, is used to regulate the temperature. The size distribution of coal and biomass is in the range of 0–0.2 mm. The bed thickness is approximately 5–10 mm at steady condition. Before the preparation of ash samples, pulverized coal and cornstalk are blended into different required ratios. Then 20–30 g pulverized blending fuel is heated to 573 K and kept for 30 min in the resistance furnace at air condition. Subsequently, the blending fuel is heated to 1073 K or 1273 K with the heating rate of 10 K/min and kept for 1 h to obtain ash samples. The ash content is calculated according to the mass of fuel and ash. Finally, ash samples are analyzed by ICP-AES to measure the content of Ca, K, Na, S, and P in ash. X-ray diffraction (XRD) is used to detect the sorts of the compounds.

2.3. Calculating methods

Chemical equilibrium analysis is widely used to predict chemical forms at thermodynamic stable state during combustion. The composition is calculated by minimizing the total Gibbs free energy in the combustion system [33]. Thermodynamic equilibrium implies the minimum total Gibbs free energy, where homogeneous and heterogeneous reactions all reach equilibrium. In this paper, the equilibrium analysis software FactSage is used to analyze the release of alkali metals during co-firing cornstalk derived fuel and coal. Based on its large database, all the elements will be considered into the calculation. After entering parameters (elementary composition of fuel and air, temperature and pressure etc.), FactSage will search the corresponding species and elements from the database automatically. Then the thermodynamic equilibrium calculation is conducted for the system with these species. The elements C, H, O, N, S, Cl, Si, P, Ca, K, Na, Mg, Al, Fe, and Ti are used to present the original fuel. 600 species (143 gas, 87 liquid and 370 solid species) are selected to conduct the equilibrium analysis calculation. The temperature range is between 400 and 2000 K, and the calculated point is set every 50 K. Although FactSage is powerful in predicting stable species during chemical equilibrium process, it still has some deficiencies in combustion calculation [33,34]. For example, the reaction time at practical condition is not long enough to reach thermodynamic equilibrium. In general, FactSage can only be used to give the equilibrium distribution of elements and the reaction mechanism of species.

3. Results and discussion

3.1. Main compounds in ash

XRD is used to analyze the main compounds in ash produced by co-firing cornstalk derived fuel and coal (Fig. 1). Alkali metals in coal ash occur as KAlSi_3O_8 , $\text{NaAlSi}_3\text{O}_8$, and Na_2SO_4 . And they occur in cornstalk ash as KFeO_2 , $\text{K}_6\text{Fe}_2\text{O}_5$, $\text{K}_2\text{S}_2\text{O}_5$, NaCl , and Na_3PO_4 .

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