



Effect of atmosphere on the release behavior of alkali and alkaline earth metals during coal oxy-fuel combustion



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HIGHLIGHTS

- Effect of atmosphere on the release of AAEM was investigated on drop tube reactor.
- Conditions that favor coal combustion promoted the release of AAEM.
- SO₂ in the atmosphere suppressed the release of AAEM.
- The presence of H₂O in the atmosphere promoted the release of AAEM.

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ABSTRACT

Understanding the effect of the atmosphere on the release of alkali and alkaline-earth metals (AAEM) during oxy-fuel combustion is important for the development of oxy-fuel combustion technologies with reduced problems with particulate formation and corrosion. To this end, we examined the release behavior of AAEM during oxy-fuel combustion in different gas environments. A drop-tube reactor and ion chromatography were used to investigate the release rates of AAEM. Our results show that the release rates of Na and K were higher than those of Ca and Mg in all types of gas environments. Experimental conditions that favor combustion, namely, higher temperature, higher oxygen content, and longer residence time, markedly increased the release rates of Na and K, whereas changes in the release rates of Ca and Mg were much smaller. The presence of SO₂ in the atmosphere suppressed the release of AAEM via formation of sulfates, whereas the presence of H₂O promoted it. A synergistic effect of H₂O and SO₂ on the release of AAEM during oxy-fuel combustion was observed.

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1. Introduction

Coal contains small amounts of alkali and alkaline-earth metals (AAEM). A significant fraction of AAEM compounds are vaporized during combustion or gasification. Although these compounds are nontoxic, they can cause a variety of problems, including fouling, slagging, and corrosion. Furthermore, recent studies [1–8] indicated that AAEM compounds contribute heavily to formation of fine particulate matter and deactivate catalysts used in flue-gas cleaning units. To address these problems, many studies have focused on the release behavior of AAEM during combustion or gasification.

Many factors [9–14], including coal properties and reaction conditions, influence the release behavior of AAEM during coal combustion or gasification. For instance, the levels and distribution of aluminosilicate and chlorine affect the release of AAEM. Specifically, a reaction between sodium chloride or potassium chloride and aluminosilicate may form a more stable product, sodium aluminosilicate, suppressing the release of AAEM. The tendency of Na and K for release initially increases and then decreases with increasing Ca/S ratio. The amount of AAEM released markedly increases with the increase in temperature. Increasing pressure suppresses the release of Na as Na carboxylates, as well as metallic Na, X–Na (X represents fragment radicals), or both. Increasing the air/coal ratio decreases the release of alkali-containing compounds in the vapor phase and increases the amount of K- and Na-containing aluminosilicates in ash. In addition to the factors mentioned above, particle size and residence time are also parameters that need to be considered. Larger particle size may prolong the diffusion time of

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AAEM and decrease release rates. Longer residence time favors the release of AAEM.

Oxy-fuel combustion is one of the most promising technologies for carbon capture and storage. It uses pure oxygen and a fraction of the recycled flue gas, which consists mainly of CO₂ and O₂. H₂O and SO₂ are concentrated via recirculation of the flue gas. Compared with traditional combustion in air, oxy-fuel combustion brings about large changes in the furnace atmosphere. This change in atmosphere leads to changes in the reaction of coal particles. Many investigations have been focused on the effect of changes in the oxidant on the oxy-fuel combustion process and on formation and control of pollutants. These changes include ignition and burnout of pulverized coal as well as heat transfer in the furnace [15–25]. Nevertheless, few studies have been conducted on the release of AAEM during coal combustion under an SO₂/O₂/CO₂ or H₂O/O₂/CO₂ atmosphere.

Both CO₂ and SO₂ can react with AAEM compounds to form carbonate and sulfate. The presence of high concentrations of steam in the system also strongly influence the char structure, thereby altering char activities [26,27] and AAEM release behavior. Therefore, we aimed to find the effect of the atmosphere on AAEM release during coal oxy-fuel combustion in the present study.

2. Materials and methods

2.1. Coal samples

A bituminous coal supplied by Yanzhou Coal Mine in Shandong Province in China, labeled as “YZ coal”, was used in the study. The coal was crushed and sieved to obtain a sample with particle sizes between 65 and 100 μm, which was used in most of the experiments. A few experiments were conducted with particle size of 100–150 μm for the purpose of comparing. The properties and AAEM concentrations of the sample are presented in Table 1.

2.2. Combustion

Combustion experiments were conducted by using a drop-tube reactor (DTR) with a quartz tube of 80 cm isothermal zone length. A schematic diagram of the setup is shown in Fig. 1. Coal particles were fed by an injector at a total gas flow rate of 600 mL/min into the reactor heated to the required temperature. An accurately weighed amount of coal (~500 mg) was fed into the reactor. Introduction of the feed required ~15 min. Coal particles fed into the reactor were heated at rates exceeding 10³–10⁴ °C/s, and the residence time in the reactor was 0.6 s. The calculation method may be found in Ref. [28].

Char was collected in a bottle for further analysis. Char yield is obtained from Eq. (1)

$$\text{Char yield} = \text{mass of char} / \text{mass of coal input} \quad (1)$$

The mass of char is determined by weighing the reactor and char-collection bottle before and after an experiment.

A fixed-bed reactor (FBR) constructed with a quartz tube was also used in several experiments. A quartz frit installed inside the reactor was located within the isothermal zone inside the electric furnace. Accurately weighed samples of coal were preloaded on

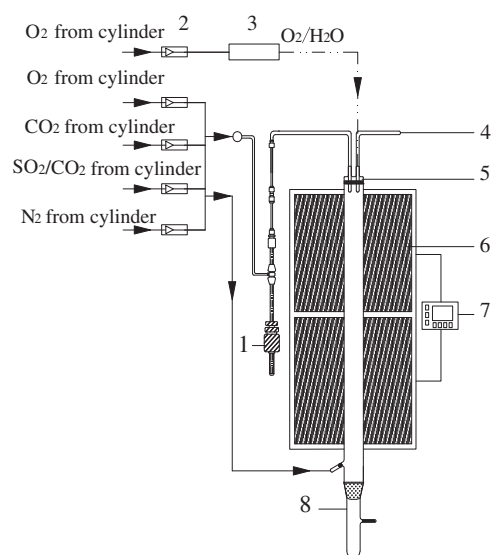


Fig. 1. Schematic diagram of the experiment setup. (1) Sample injector; (2) Flow controller; (3) Steam generator; (4) Gas outlet; (5) Drop-tube reactor; (6) Electric furnace; (7) Temperature controller; (8) Sample-collection bottle.

the quartz frit of the reactor. After the air in the tube was replaced with the reaction atmosphere at a flow rate of 400 mL/min, the reactor was heated at a rate of 10 °C/min to the specified temperature. The temperature was then maintained for 30 min. Subsequently, the char was cooled in nitrogen, collected, and then weighed for further analysis.

O₂, CO₂, and SO₂ stored in cylinders were used. Different concentrations of O₂ could be obtained by adjusting the flow rates of O₂ and CO₂. The SO₂ concentration in the atmosphere used in this study was 7300 ppm. H₂O was introduced into the drop-tube reactor by entrainment of saturated steam at 82 °C. Finally, an atmosphere containing 10% (v/v) steam was obtained. All concentrations of gases mentioned in this work are volume percentages, unless otherwise indicated.

2.3. Quantification of Na, K, Mg, and Ca

Concentration of Na, K, Mg, and Ca were analyzed by ion chromatography (IC). The sample for IC analysis was prepared as follows. Accurately weighed portions of coal particles or char particles were placed into a cupel and then ashed according to the method GB/T212-2008. The ash was then placed in a Teflon vial for digestion in a HClO₄/HF solution (1:3, v/v, analytical grade; Tianjin Chemical Reagent Research Institute) at 110 °C for 3 h. After digestion, the acid-digested ash was dissolved in 0.02 M methanesulfonic acid (Aladdin Reagents (Shanghai) Co. Ltd., Guaranteed reagent). Quantification of Na, K, Mg, and Ca content was carried out by an ion chromatography using a CSRS cation suppressor and a CS12 column.

The release rate of AAEM was calculated by considering the amounts of both char and raw coal, as well as the yields of char

Table 1
Proximate and Ultimate analysis of YZ coal.

Sample	Proximate analysis (wt.%)			Ultimate analysis (wt.%, daf)					AAEM concentration (wt.%, ad)			
	M _{ad}	A _d	V _d	C	H	N	S	O*	Na	K	Mg	Ca
YZ	2.29	13.63	43.33	81.54	5.07	1.29	5.00	7.10	0.28	0.09	0.06	0.83

* By difference.

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