Powder Technology 302 (2016) 328–332

Contents lists available at ScienceDirect

Powder Technology

journal homepage: www.elsevier.com/locate/powtec

Short communication

Release behavior of Hg, Se, Cr and As during oxy-fuel combustion using Loy Yang brown coal in a bench-scale fluidized bed unit

Bithi Roy [a,b,*], Sankar Bhattacharya [a]

[a] Department of Chemical Engineering, Monash University, PO Box 36, Clayton Campus, Victoria 3800, Australia
[b] Currently at School of Chemical Engineering, University of Adelaide, Adelaide, South Australia 5005, Australia

ARTICLE INFO

Article history:
Received 22 April 2016
Received in revised form 28 August 2016
Accepted 30 August 2016
Available online 31 August 2016

Keywords:
Brown coal
Fluidized bed
Mercury
Oxy-fuel
Trace elements

ABSTRACT

This experimental study investigates, for the first time, the fate of trace elements (Hg, Se, Cr and As) during combustion of Loy Yang brown coal in a large bench-scale oxy-fuel fluidized bed. The results indicated that up to 70%, 28%, 7% and 2% of total As, Cr, Se, and Hg respectively were retained in the collected ash at 15–30% oxygen concentration in the oxy-fuel combustion atmosphere at around 850 °C. With the increase in oxygen concentration in oxidant, the Hg adsorption in ash was found to decrease. Compared to bed ashes, higher Hg adsorption was observed in fly ashes containing higher carbon content. Therefore, a higher level of Hg capture is expected in oxy-fuel circulating fluidized bed combustion due to the circulating materials having higher carbon content.

1. Introduction

Oxy-fuel fluidized bed (Oxy-FB) combustion has been attracting growing attention due to its potential to reduce CO_2 emission from power plants handling a wide variety of coals. Moreover, fuel flexibility, low NO_X emission due to low operating temperature and low SO_2 emission via in-bed sorbent addition are the known advantages of Oxy-FB combustion [1]. However, the issue of trace elements emissions during coal combustion has an important implication on the operation of Oxy-FB combustors using different types of coals. The fate of trace elements, which are present in coal at low concentrations of below 100 ppm [2], are an important consideration as the inclusion of excessive amounts of these elements beyond the permissible limits is harmful to the health and environment [3–5], with additional implications for CO_2 transport and storage [6]. Furthermore, the trace elements emission characteristics are of great importance for the design of gas cleaning systems for carbon capture and storage purposes.

During coal combustion, mercury (Hg), selenium (Se), arsenic (As) and chromium (Cr) are considered as the elements of greatest environmental and health concern [2]. Though several studies have been carried out to investigate the distribution of trace elements in air-fired fluidized bed combustion [7–9], limited experimental data are available on oxy-fuel fluidized bed combustion [10–12]. Therefore, it is important to know the release behavior of trace elements during Oxy-FB combustion using a wide range of coals including brown coals. Australia has a large resource of brown coal, over 500 years at the current rate of consumption [11]. Using three Australian brown coals - Loy Yang, Morwell, and Yallourn, in our previous study [13], the speciation of chromium, arsenic, selenium and mercury was predicted by thermodynamic equilibrium modeling under oxy-fuel combustion condition. This modeling study, however, recommended generating experimental data to know the actual distribution of these elements under practical operating conditions.

This experimental study investigates, for the first time, the fate of these trace elements using Loy Yang coal in a 10 kW_{th} fluidized bed combustor under oxy-fuel combustion. The effects of oxygen and steam in combustion atmosphere on the trace elements retention in ash are also considered in this study. Additionally, the results from oxy-fuel combustion are compared to those from air combustion.

2. Materials and methods

2.1. Feedstock

Experiments were carried out with one air-dried Australian brown coal - Loy Yang with a starting particle size of 1–3 mm. The composition of the coal is given in Table 1. Silica sand having a particle size of 350–400 μm was used in the reactor as a bed material. Moreover, char (77.97% C, 2.42% H, 1.29% N, 0.11% S, 15.77% O and 2.44% ash) with the particle size of 1–3 mm was used during start-up. All char was used up during the heat-up period, as indicated by commissioning run using only char as feedstock.

* Corresponding author.
E-mail address: bithi.roy@monash.edu (B. Roy).

http://dx.doi.org/10.1016/j.powtec.2016.08.073
0032-5910/

Table 1
Composition of the Loy Yang coal used in experiments.

Ultimate analysis (wt.% dry basis)		Minerals and inorganic (wt.% ash basis)		Trace element (mg/kg dry coal basis)	
Carbon	65.00	SiO_2	56.90	Cr	16.00
Hydrogen	4.60	Al_2O_3	20.64	As	0.30
Nitrogen	0.72	Fe_2O_3	4.63	Se	1.00
Sulphur	0.50	TiO_2	1.51	Hg	0.06
Oxygen	25.48	K_2O	1.31		
Ash	3.70	MgO	3.63		
		Na_2O	4.73		
Chlorine	0.11	CaO	1.61		
		SO_3	5.04		

2.2. Experimental installation

The experimental installation mainly consists of a bench-scale 10 kW_{th} fluidized bed combustor, electric furnace, screw feeder, gas supply unit, gas pre-heating system, and cyclone separators. The combustor is of 0.1 m diameter and 1.5 m length, and the freeboard of 0.15 m diameter and 2 m length. To simulate typical feed gas composition under oxy-fuel combustion conditions, all gases - O_2, CO_2, and steam, were mixed and pre-heated before entering into the reactor. A detailed description of the experimental installation was given in our previous studies [12,14]. Experimental conditions are summarized in Table 2. It is noted that during experiments the inlet temperatures at primary cyclone and secondary cyclones were around 410 °C and 200 °C respectively with ±20 °C variation.

After each experiment, the fly ash, and bed ash samples were collected from two (primary and secondary) cyclone pots and fluidized bed respectively for trace elements analysis. Both arsenic and selenium were analyzed according to the standard method AS 1038.10.2, whereas chromium and mercury were determined according to the standard methods AS 1038.10.1 and AS 1038.10.5 respectively. All analyses were repeated minimum three times, and the average values are reported in this paper.

3. Result and discussion

3.1. Chromium

It can be seen from Fig. 1 that compared to air combustion, the extent of chromium in total ash was lower (by 22%) in oxy-fuel combustion even at the same oxygen concentration (21% v/v). Moreover, with an increase in O_2 concentration in feed gas under oxy-fuel combustion, the concentration of chromium was observed to decrease slightly in ashes. The Cr retention decreased by 7% when the oxygen concentration was increased from 15% (v/v) to 30% (v/v) in the combustion atmosphere. This is because a higher level of oxygen concentration is known to help to vaporize the mobile Cr from char surface, consequently resulting in lower Cr retention in ash [15].

For Loy Yang coal, it is also evident that steam in the combustion atmosphere did not affect the Cr vaporization, as shown in Fig. 1. This result, however, is different from the study by Wang et al. [15], who found lower Cr retention in the presence of steam under oxy-fuel combustion atmosphere using Chinese bituminous coal. As shown in our previous study [13] on thermodynamic prediction, it can be said that Cr retention is dependent on coal composition to some extent.

The experimental results indicated that maximum 28% of total coal-Cr was retained in the total collected ash under oxy-fuel combustion. Since the low volatile Cr is mostly captured in fly ash, it should be kept in mind that the Cr capture in the fly ash could result in the erosion of alloy steel or stainless steel gas sampling probes in the combustor [16].

3.2. Arsenic

In the case of arsenic retention, no significant difference was observed between air combustion and oxy-fuel combustion at the same oxygen concentration of 21% (v/v), as indicated in Fig. 2. However, the arsenic adsorption in ash was found to decrease with increase in oxygen concentration in the feed gas. When the oxygen concentration was increased from 15% (v/v) to 30% (v/v) in the combustion atmosphere, arsenic retention decreased by 13%. This finding is consistent with the observations by other researchers [15], who also observed lower As concentration in fine ashes at higher oxygen concentration under oxy-fuel combustion.

The experimental results also showed that the addition of steam (even with same 15% v/v oxygen concentration in oxidant) in the combustion environment promoted (by 11.5%) the arsenic retention. Several researchers [15] also observed the greater extent of total As in particulate matter during oxy-fuel combustion attributing this to the fact that H_2O enhances the formation of particulate matter, which in turn results in higher arsenic retention.

From the experimental results, maximum 70% of arsenic in coal was found to remain in ash. However, further analysis is recommended to identify the proportion of water leachable arsenic in the ash sample from the disability aspect of ash samples.

3.3. Selenium

From Fig. 3 it can be seen that compared to air combustion, Se retention in total ash was significantly lower (by 60%) in oxy-fuel combustion at the same oxygen concentration. Around 17% of total coal-Se was observed to retain in the collected ash in air combustion, whereas in oxy-fuel combustion only 7% of total coal-Se was found to adsorb in the collected ash. With the variation of oxygen concentration in the feed gas, however, no systematic trend was observed in the experimental results, as shown in Fig. 3. This finding is consistent with the observation by Contreras et al. [17] using sub-bituminous coal, who also did not find any difference in Se volatilization with an increase in oxygen concentration in the O_2/CO_2 mixture. They concluded that in oxy-fuel combustion, the trace elements behavior was highly dependent on the fuel composition.

Moreover, it was noticed that the addition of steam with the same 15% (v/v) oxygen concentration in feed gas lowered (by 15%) the selenium retention in ash. This is because steam can promote the

Table 2
Operating conditions used in experiments.

Combustion atmosphere (% volume)	Coal feed rate (g/h)	Gas velocity (m/s)	Bed temperature (°C)	Steady-state period (h)	Carbon content in fly ash (%)	
					Primary cyclone ash	Secondary cyclone ash
Air	650	0.80	840 ± 10	4.50	1.12	0.27
15% O_2 + 85% CO_2	850	0.80	840 ± 10	3.25	1.39	0.45
21% O_2 + 79% CO_2	850	0.80	840 ± 10	3.50	0.66	0.28
30% O_2 + 70% CO_2	800	0.80	845 ± 5	3.75	0.77	0.18
15% O_2 + 73% CO_2 + 12% H_2O	840	0.75	860 ± 10	2.00	1.63	0.55