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Partitioning behavior of trace elements in a 2.5MW$_{th}$ pilot circulating fluidized bed combustor burning anthracite and bituminous coal

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Abstract: Coal, as the major energy resource in countries like China and India, introduces large amounts of pollutants into the atmosphere, including trace elements originally bonded to the coal matrix. These trace elements pose an environmental and human health risk, to which more attention needs to be paid, but few studies have been done, particularly for circulating fluidized beds (CFB). This paper discusses tests with anthracite and bituminous coal combustion carried out in a 2.5MWth CFB combustor with multi-stage control of solids. The partitioning of seven elements (As, Ba, Cd, Cr, Cu, Mn and Pb) into different solid streams was investigated. The mass balance ratio of the studied elements ranges from 56%-137%, which is, with respect to their concentrations, satisfactory and reasonable. Most of the elements were found in bottom ash and fly ash during CFB combustion, while small amounts of As, Cd and Pb were emitted into the atmosphere with fine particulates. The trace elements are more likely to be retained in bottom ash when burning bituminous coal than when burning anthracite. For the volatile elements, the enrichment in solid streams follows the trend of: bag filter ash > cyclone ash > IBHX (In-bed heat exchanger) solids > bottom ash, which confirms the expectation that the volatile elements tend to

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be enriched in fine particles. Anthracite, compared to bituminous coal, shows lower emission factors for all monitored elements, except for Pb, which has a higher emission factor during anthracite combustion. This study should serve as a good reference for trace element control strategies in coal-fired CFB boilers.

Key words: Trace elements; circulating fluidized bed; mass balance ratio; enrichment; emission factor

1. Introduction

Coal combustion is one major source of pollutants in the atmosphere. During the coal combustion process, some trace elements bonded with the coal's mineral and organic matter components will be released and may pose an environmental and human health risk depending on their concentration, toxicity and partitioning behavior in the combustion and environmental control systems [1-3].

Generally, the trace elements that are not volatilized during combustion will be retained in the coal ash, while the volatile elements will mostly vaporize. As the flue gas flows through the downstream sections of the boiler, some elements will condense on the surface of particles and will recombine with the particulate by physical adsorption or chemical reaction at low temperature. The trace elements absorbed on particles will be collected by the flue gas clean-up devices, such as wet scrubbers, ESPs and fabric filters, while the most volatile elements will be emitted into the atmosphere. The final distribution of the trace elements into different product streams is strongly influenced by their occurrence in the coal matrix, the combustion temperature, the configuration of the furnace and the gas clean-up devices.

Circulating fluidized bed (CFB) combustion has been recognized as an important clean coal technology due to its use of low temperature combustion (normally around 850℃) and good emission control of SO_2 and NO_x by injecting limestone and by air staging, respectively. Since there are big differences in combustion temperature, ash split and flue gas clean-up devices between pulverized coal (PC) combustion and CFB combustion, the trace elements partitioning behavior should also be different. For example, the PC furnace temperature is usually around 1300℃ and more elements will experience the vaporization-condensation process, so there will be greater enrichment of trace elements in fine particles. Due to the hydrodynamic differences, the ash during PC combustion is mainly released as fly ash, while, during CFB combustion, the fly ash and bottom ash ratio varies over a wide range from 2:8 (when burning coal waste) to 9:1 (when burning coal sludge). Furthermore, PC boilers are usually equipped with a wet flue gas desulfurization unit (WFGD), while, in CFB boilers, the desulfurization takes place inside the furnace by means of limestone injection. All of these differences will affect the trace element transformation processes. However, there is little published on trace element partitioning behavior in CFBs compared with PC boilers [4-5]. Further, trace element studies in fluidized beds have focused mainly on emissions when burning or gasifying fuels like biomass and municipal solid waste rather than coal [6-11]. But since there are huge amounts of coal used in fluidized beds in countries like China and India, it is also very important to study the trace element partitioning behavior during coal combustion in CFBs. Although there are papers on element partitioning during bituminous coal and lignite combustion in fluidized beds [12-14], this study is the first one to report the trace element partitioning behavior during anthracite combustion in a fluidized bed.

In this study, one anthracite and one bituminous coal were combusted in a 2.5MW_{th} CFB

combustor with multi-stage solid control system. The distribution of trace elements (As, Ba, Cd, Cr, Cu, Mn and Pb) in various solid streams was studied. The results will serve as a reference on trace element partitioning behavior during coal combustion in CFBs.

2. Experimental

2.1 Fuel analysis

Two typical Chinese coals, one anthracite and one bituminous coal, were used to carry out the experimental program. The ultimate and proximate analyses of the coal samples are provided in Table 1. The coal samples were crushed to produce particles with a maximum diameter of 6mm. The particle size distribution is shown in Fig. 1. The Sauter Mean Diameters (SMDs) are calculated as 0.51 mm and 0.61mm for anthracite and bituminous coal, respectively.

The trace element content in the samples was analyzed by inductively-coupled-plasma mass spectrometer (ICP-MS, 7700x, Agilent, USA) after the acid digestion procedures described in reference 13. The results for the seven trace elements, Arsenic (As), Barium (Ca), Cadmium (Cd), Chromium (Cr), Copper (Cu), Manganese (Mn) and Lead (Pb), as well as the detection limit of the ICP-MS are shown in Table 2. Some other trace elements, Beryllium (Be), Cobalt (Co), Molybdenum (Mo) and Antimony (Sb), were also measured, but the results are not included here, since the content in the two samples was low and high accuracy could not be guaranteed. The Ba content is higher than 100ppm and so cannot be strictly named as a trace element, but the results are included here since it is usually treated as trace element when burning other fuels. The ash composition of the two coal samples was also analysed by X-ray fluorescence (ARL9800XP, Thermoscientific, USA). The results are presented in Table 3.

2.2 Facility description and operation

Each test was carried out in a pilot CFB combustor with a heat input of 2.5MW. The system is shown schematically in Fig.2. There are several critical characteristics of the system. First, two stages of solid separation are involved including an in-furnace U-beam separator operated at high temperature, around 850℃, and a downstream cyclone separator operated at medium temperature, from 350℃ to 450℃. Second, there is an in-bed heat exchanger (IBHX) installed parallel to the riser which allows exchange of solids between it and the main riser and approximately 16% of the total heat load can be removed by the heat surface in the heat-exchanger. Third, the system is equipped with an advanced operating control and data acquisition system, based on a programmable logic controller (PLC) platform, which can facilitate the operation very conveniently.

In terms of operation, the heat input was strictly controlled at 2.5MW_{th} by fixing the coal-feeding rate as constant. The bed temperature of the CFB was controlled at 880±20℃. The primary air ratio was kept in the range of 60%-65%. The excess oxygen concentration before the bag filter was controlled at 3.5%-4.2%, with the excess air coefficient at 1.20-1.25. The gas temperature in the bag house was around 150℃ to avoid the condensation of moisture. The bed material was the conditioned bottom ash from the same coal combustion for each test, with a size range of 1-3 mm. No limestone was injected into the furnace during these tests.

2.3 Sample collection

After the operation was stabilized for more than 8 h, which means the solid flow of each loop was established and remained steady, the solid sampling was carried out. The bottom ash of the riser

was drained and collected from the bed drain duct after purging the cold material accumulated in the drain duct, to make sure the drained solids were indeed the hot bed ash produced under the appropriate experimental conditions. The fly ash was collected at the discharge hopper of the bag filter. The discharge of fly ash was continuous, so the fly ash was considered as being well conditioned. The solids captured by the cyclone were collected from the loop-seal exit, which is marked as ③ in Fig.2. The IBHX bed solids were sampled by a specially designed in-bed probe to extract the solids from an open port in the back wall of the IHBX, which is shown as ④ in Fig.2. Two samples were collected from each location every half hour and then well mixed for elemental analysis. Each mixed sample was split equally into 4 samples and one of them was tested twice to ensure the reproducibility of trace element measurement. The concentration of trace elements in the flue gas was not measured in the test, since all the trace elements, except for the very volatile ones like Hg (which is not considered here), were assumed to be completely condensed and recombined with the ash when the flue gas temperature dropped down to 150 ℃.

2.4 Sample analysis

The unburnt carbon content in all ash samples was analysed by the measurement of weight loss after heating the samples in a muffle furnace for 7 minutes at 900 ℃. A microwave dissolver (MDS-6, Zhongnuo Chemical Technology, China) was used to acid digest the ash sample according to EPA Method 3050B. Then the content of the same seven elements was analysed using the ICP-MS (7700x, Agilent, USA).

3. Results and discussion

3.1 Mass balance of the trace elements

After the combustion is stabilized, the elements introduced into the furnace will exit the system in three streams: bottom ash, fly ash and flue gas into the stack. The total mass balance can be expressed by the following equation:

$$M_{oi} = M_{bi} + M_{fi} + M_{gi} \quad (1)$$

Where M_{oi} is the total amount of element i introduced into the furnace within coal, mg/h; M_{bi} is the amount of element i discharged with the bottom ash, mg/h; M_{fi} is the amount of element i leaving the system with the fly ash, mg/h; and M_{gi} is the amount of element i leaving the system with flue gas, mg/h.

The trace elements in the gas phase can be collected as described in EPA Method 29. However, as pointed out in the experimental section, nearly all the elements in the gas were considered as being re-condensed on the solids, so the parameter M_{gi} can be neglected in this study. This assumption has already been validated by many previous studies [6-10], which found that nearly all the elements are present in the ash stream rather than in the flue gas.

The ash split into bottom ash and fly ash is usually not easy to determine in CFB combustion because it continues to change as the operating conditions change. In this study, the ash recovery rate was closed at 97% based on the long period of operation and the bottom ash/fly ash ratio was calibrated as 4:6 by weighing the purged bottom ash and fly ash separately. Also, the elements carried by the fines into the stack were not taken into account due to the very high capture efficiency (>99%) of the bag filter. In consequence, the total of all elements leaving the system M_{hi} can be expressed as:

$$M_{hi} = M_{bi} + M_{fi} = \left(0.4 * \frac{C_{bi}}{A_b} + 0.6 * \frac{C_{fi}}{A_f}\right) \cdot M \cdot A \quad (2)$$

Where M_{hi} is the total amount of element i leaving the system, mg/h; C_{bi} and C_{fi} are the concentrations of element i in the bottom ash and fly ash, mg/kg; A_b and A_f are the ash contents in the bottom ash and fly ash, which is 1 minus the unburnt carbon content in the ash; M is the coal feeding rate, kg/h; A is the ash content in the original coal.

The unburnt carbon content in the bottom ash for both anthracite and bituminous coal is less than 1%. The unburnt carbon content in the fly ash is 22.6±0.8% for anthracite and 12.7±0.5% for bituminous coal. Anthracite is more difficult to burn out completely in the CFB combustor due to its lower volatiles and higher fixed carbon. By calculating the M_{hi}/M_{oi}, a picture of the mass balance ratio of each element can be obtained, as shown in Fig.3.

As can be seen from Fig.3, for anthracite, the mass balance ratio for all the elements ranges from 77%-115%, except for Mn and Cd, which are 131% and 56%, respectively.; For bituminous coal, the mass balance ratio for all the elements ranges from 85%-122%, except for Cr and Mn, which are 137% and 134%, respectively. The results are consistent with the results from previous studies. Selcuk et al [15] investigated element partitioning behavior by burning coal in a 0.3 MW_{th} fluidized bed and found the mass balance of Cd, As and Mn to be 25%-45%, 95%-175% and 150%-250%, respectively. Reed et al [16] argued that the mass balance ratio of 100±30% is normally satisfactory for trace element studies in pilot gasifier plants. Åmand and Leckner [6] found the recovery ratio of different elements varies from less than 10% to more than 200% when burning sewage sludge and coal/wood in their 12 MW_{th} CFB boiler. In circulating fluidized bed combustion, a complete closure of element mass balance is very difficult to achieve because a certain amount of solids of the proper size will be captured by the cyclone and accumulate in the furnace rather than purging out, and the trace elements combined with these particles will not leave the system particularly in the start-up periods. Furthermore, the element distributions in both the coal being fed and the ash product are not completely uniform themselves. Also, there is always a measurement error with the ICP-MS. Nonetheless, the results in this study are reasonable.

3.2 Trace element partitioning in bottom ash and fly ash

By quantifying the M_{bi}/M_{hi} and M_{fi}/M_{hi}, the partitioning of each trace element in the bottom ash and fly ash is shown in Fig.4.

During anthracite combustion, most of the trace elements are present in the fly ash, which takes up 80%-90% of the M_{hi}. Only 10%-20% of the elements are retained in the bottom ash, where Pb is the most enriched metallic element. When burning bituminous coal, roughly half of the Ba, Cr and Mn are retained in the bottom ash, while all other elements in the bottom ash are no more than 30%. More than 80% of Cd is present in the fly ash, which is the highest share in fly ash for bituminous coal.

For both coals, the mass balance ratios of As, Cd and Pb are all less than 100%, as shown in Fig.3, which indicates these three volatile elements may be emitted into the atmosphere along with the fine particles from the bag filter. Bunt and Waanders [17] consider these three elements to be volatile ones that are prone to be present in the flue gas. Syc et al. [18] found 80%–90% of Cd will not condense at temperatures higher than 380℃ in a fluidized bed biomass gasifier. This may cause the Cd to be emitted into the atmosphere before complete condensation. The presence of $CdCl_2$ in the gas phase in an 850℃ fluidized bed was also verified by both experimental and model study [19]. It is also well known [20] that As will be present mostly in the gas phase during combustion and as the temperature falls, a substantial amount of it will still be present in the gas phase. For both coals, about 80% of the Cu is present in the fly ash. In anthracite, the Cu yield is less than 100%, which indicates the Cu also has some volatility, but obviously this is weaker than that observed for As, Cd and Pb. The mass balance ratios of Ba, Mn and Cr are all higher than

100% and most of them are present in the bottom ash, which confirms that they are all non-volatile at the CFB combustion temperature.

The partitioning of trace elements depends very much on the coal type, as shown in Fig.4. The trace elements in bituminous coal are more likely to be present in bottom ash, which may be caused by the differences of element occurrence and ash composition between bituminous coal and anthracite. The case for As is more easily verified because several studies have been done on As transformation during combustion. In this study, 14% of As goes into the bottom ash when burning anthracite and 30% when burning bituminous coal. A previous study [21] revealed that the As bonded with the organic sulfur is vaporized and then condensed on the surface of the fine particles while the As bonded with the scattering minerals is retained in the ash residues. As shown in Table 3, the ash of bituminous coal has about 20% CaO, which has a clear effect on its absorption ability towards As. The following reaction during combustion will definitely help to retain the As in the bottom ash of bituminous coal as the product is quite difficult to vaporize.

$$3CaO+As_2O_3 \rightarrow Ca_3(AsO_3)_2 \quad (3)$$

3.3 Relative enrichment factor of trace elements in different solid streams

Due to the large variation in specific element concentration in different coals, it is better to use the relative enrichment factor, which normalizes the enrichment behavior of different elements in different coals. The relative enrichment factor proposed by Meij et al [22] is often used to describe the relative enrichment behaviors. Taking the relative enrichment factor of element i in the fly ash as an example, this can be expressed as:

$$RE_i = \frac{C_{fi}}{C_{oi}} \cdot A \quad (4)$$

Where RE_i is the relative enrichment factor of element i in the fly ash, C_{fi} is the concentration of element i in the fly ash, C_{oi} is the concentration of element i in the original coal, and A is the ash content in the original coal.

For this definition of relative enrichment, only the ash content in the original coal is considered while the ash content in the solid product is neglected which may cause the factor to be influenced by the unburnt carbon content in the solid product. In this paper, the relative enrichment factor is modified as

$$I_{RE} = \frac{C_{fi}}{C_{oi}} \cdot \frac{A}{A_o} \quad (5)$$

Where I_{RE} is the modified relative enrichment factor and A_o is the ash content in the solid product. If $I_{RE}=1$, the element neither concentrates nor depletes in the sample; if $I_{RE}>1$, the element tends to concentrate in the sample; if $I_{RE}<1$, the element tends to deplete in the sample.

The relative enrichment factors of the seven elements in the four different solid streams (bottom ash, IBHX solid, cyclone ash and bag filter ash) are presented in Fig.5.

For anthracite, the I_{RE} of all the seven elements in the bottom ash is less than 1, indicating all the elements tend to be depleted in the bottom ash and enriched in the fly ash. The I_{RE} of all the elements in IBHX solid is larger than bottom ash but smaller than fly ash except for the Cr which is larger than 1. For bituminous coal, the As, Cd, Cu and Pb tend to be depleted in the bottom ash and enriched in the fly ash. These four elements are also depleted in the IBHX. Ba, Mn and Cr tend to be enriched in the bottom ash, while their relative enrichment factors in fly ash are from 0.9-1.2, which are also not depleted. For these three elements, because their mass balance ratios

237 are all higher than 100%, they may be enriched either in bottom ash or fly ash. The results are to
238 some extent counter intuitive, but the following should be considered:

239 1) The volatile elements like As, Cd, and Pb as well as the semi-volatile Cu may experience a
240 vaporization and condensation process, so they can be expected to tend to be enriched in the fine
241 particles, which usually have bigger surface areas. The particle size distribution of the bottom ash
242 and the IBHX were sieved and measured, as shown in Fig.6. The SMDs of bottom ash and IBHX
243 solids for anthracite are 0.390mm and 0.277 mm, and they are 0.337mm and 0.273mm for
244 bituminous coal. At the same time, the particle size distribution of the cyclone ash and filter ash
245 were determined by a laser particle size analyzer (Mastersizer 3000, Malvern, UK). The results are
246 presented in Fig.7. The SMDs of the bag filter ash and cyclone ash for anthracite are 11.05μm and
247 23.48μm while they are 8.09μm and 26.5μm for bituminous coal. So it is easy to demonstrate that
248 the solid particle sizes in the four main four streams are: bottom ash > IBHX solid > cyclone ash >
249 bag filter ash. This sequence is in good agreement with the enrichment degree of the volatile
250 elements in them. Furthermore, the pore size, surface area and pore volume of cyclone ash and bag
251 filter ash were measured by a physisorption analyzer (ASAP 2000M, micromeritics, USA) and the
252 result in Fig.7 shows that the bag filter ash has a smaller pore size, as well as larger BET surface
253 area and BJH pore volume, which indicates the gas phase elements are more easily condensed on
254 it and absorbed by it physically and chemically.

255 2) Pb in coal will fully vaporize at around 850℃ in the form of $PbCl_4$ and $PbCl_2$ [23] and then the
256 gas phase Pb can be captured by the Al_2O_3 and SiO_2 particles as the temperature falls, which can
257 be express as the following equation [24]:

258 $$PbCl_2+Al_2O_3+2SiO_2+H_2O \rightarrow PbO\cdot Al_2O_3\cdot 2SiO_2+2HCl \quad (6)$$

As the cyclone in the study was operated at temperatures around 350℃–450℃, the Pb leaving the furnace was condensed and absorbed by the cyclone ash again. As a consequence, the Pb in anthracite is even more enriched in cyclone ash than bag filter ash.

3) The most volatile elements, like As and Cd, will be mostly enriched in the fine particles that have larger surface areas. The enrichment factor of As and Cd in bag filter ash is bigger than that in cyclone ash, except for the case of As in bituminous coal. The As in bituminous coal is slightly more enriched in cyclone ash than in bag filter ash, which is probably because the As_2O_3 in the flue gas can be captured by the high concentration of CaO in the cyclone ash by the reaction $3CaO+As_2O_3 \rightarrow Ca_3(AsO_3)_2$.

4) The non-volatile elements, like Ba, Cr and Mn, do not vaporize at CFB combustion temperatures. All these elements will stay in the ash, either retained in the bottom ash or in the fly ash, which is elutriated from the furnace. Their partitioning therefore depends on their occurrence in the coal matrix. Mn is bonded with the mineral matter in the coal. Cr will be present in the form of Cr_2O_3 during combustion and always behaves as a solid. Ba, with its most common naturally occurring minerals of barite and witherite, is also difficult to vaporize. In the study, the Ba, Mn and Cr in bituminous coal are enriched in bottom ash. However, these three elements in anthracite are all depleted in the bottom ash, which might be due to their different occurrences in the original coal matrix as well as the fragmentation property of the coal samples. Similar results can be found in reference 13 where different combinations of lignite and biomass were fired.

3.4 Emission factors of trace elements into the atmosphere

The above discussion doesn’t consider the trace elements released by the fines into the atmosphere, because it is assumed to be very low. However, if we want to evaluate the effects of the trace

element emissions on the environment, the elements emitted into the atmosphere relative to the energy consumed must be quantified. The emission factor of a certain element i is used here and defined as:

$$EF_i = (C_i/H)F_i \quad (7)$$

Where EF_i is the emission factor of elements i into the atmosphere, mg/kJ; Ci is the concentration of element i in the coal, mg/kg; H is the low heating value of coal, kJ/kg; F_i is the emission ratio of element i into the atmosphere.

For the non-volatile elements, assuming the bag filter was operated at reasonable separation efficiency, which is 0.99 here, the F_i can be calculated as:

$$F_i = 0.01 * \frac{M_{fi}}{M_{hi}} \quad (8)$$

And for the volatile elements, the Fi is calculated as the difference between 100 and the mass balance ratio in Fig.3. The results of F_i and EF_i are shown in Table 4 and Table 5.

The emission factor of each element depends on the coal type, the element content in the coal and the physical and chemical property of the element itself. It can be seen from Table 5 that the emission factors of anthracite are higher than those of bituminous coal for all elements, except Pb. The emission factors are dependent on the fine particulate emissions during combustion. In pulverized coal combustion, due to the higher combustion temperature, more elements vaporize into the gas and condense on the fine particulates. Lind et al [14] revealed that the ultrafine particle emission in CFB combustion is one to two orders of magnitude lower than that observed in PC combustion. Also, it has been reported [25] that the emission factors of Mn in PC combustion can rise to 15% while that of Cr can reach 61%, which is much higher than what was found here. Thus

it is clear from these results that CFB combustion may be superior in terms of the control of trace element emission, when compared with PC combustion.

Conclusions:

Partitioning behaviors of seven trace elements (As, Ba, Cr, Cu, Pb, Mn and Cd) were investigated in a 2.5MWth CFB combustor burning anthracite and bituminous coal, and the results show that:

1) The mass balance ratio of all the seven elements ranges from 56%-137%, and for most of them from 77%-115%.

2) Nearly all the elements are present in bottom ash and fly ash during CFB combustion, while a small amount of As, Cd and Pb is emitted into the atmosphere with fine particulate.

3) When burning bituminous coal, the trace elements are more easily enriched in the bottom ash than when burning anthracite.

4) For the volatile elements, the enrichment in solid streams follows the trend: bag filter ash > cyclone ash > IBHX solids > bottom ash, which indicates the volatile elements tend to be enriched in the fine particles.

5) Anthracite has a smaller emission factor than bituminous coal for all elements except Pb, which is the opposite.

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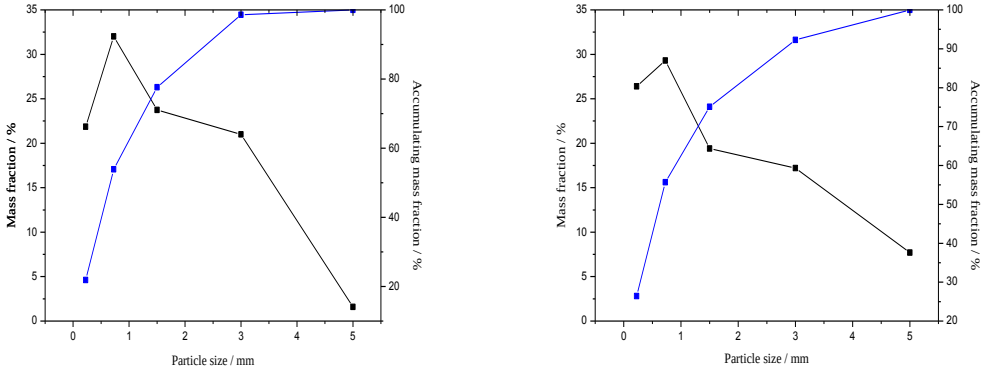
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(a) anthracite (b) bituminous coal

Figure 1 Particle size distributions of the coal samples

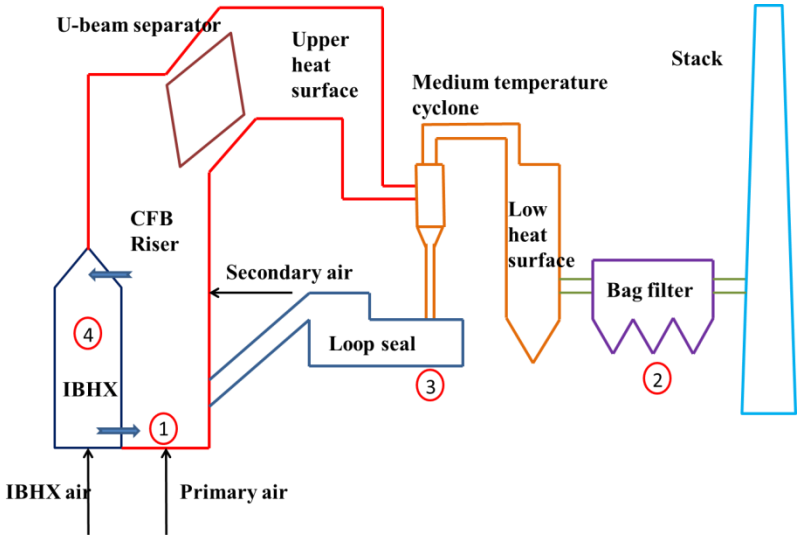
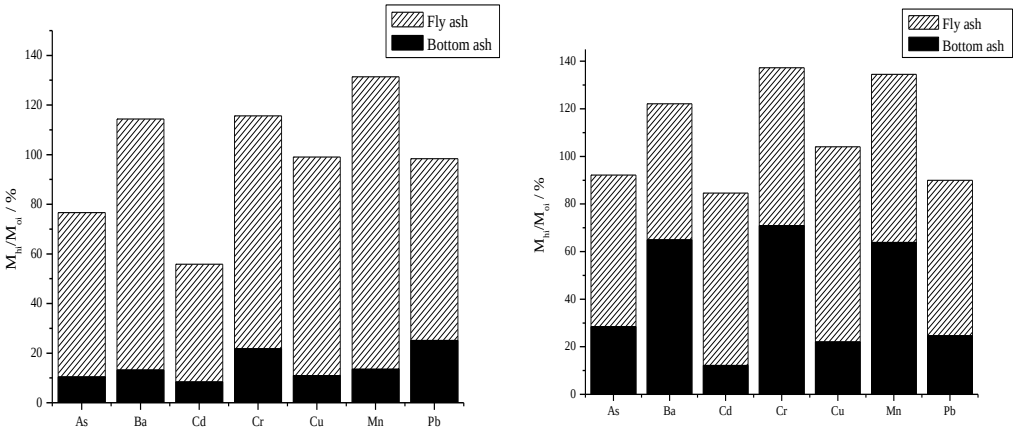
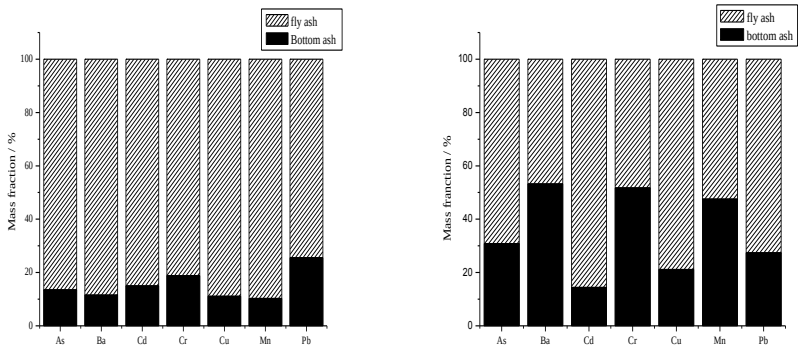


Figure 2 The 2.5 MW_{th} CFB combustion system



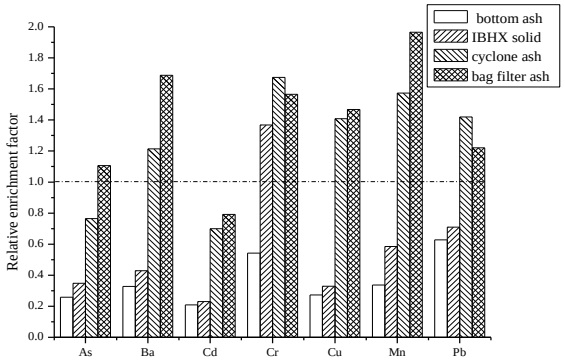
(a) anthracite (b) bituminous coal

Figure 3 Mass balance ratio of trace elements in anthracite and bituminous coal

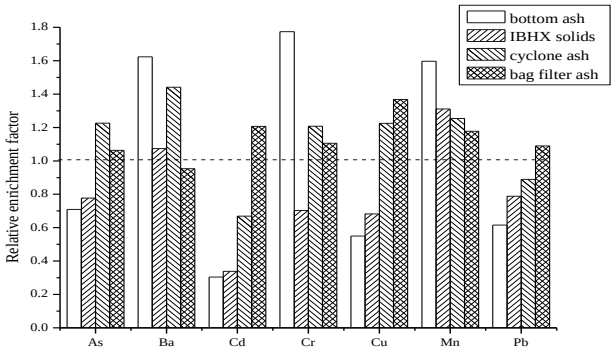


(a) anthracite (b) bituminous coal

Figure 4 trace elements partitioning into fly ash and bottom ash

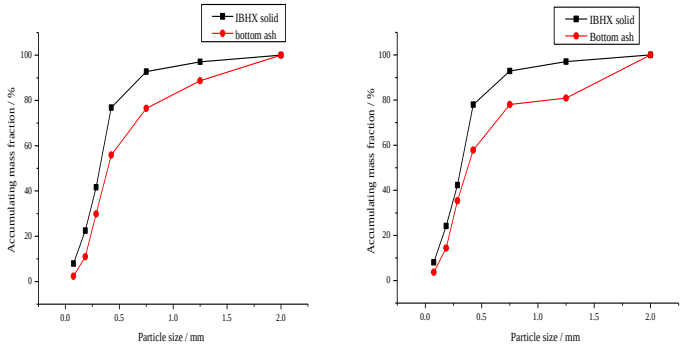


(a) anthracite



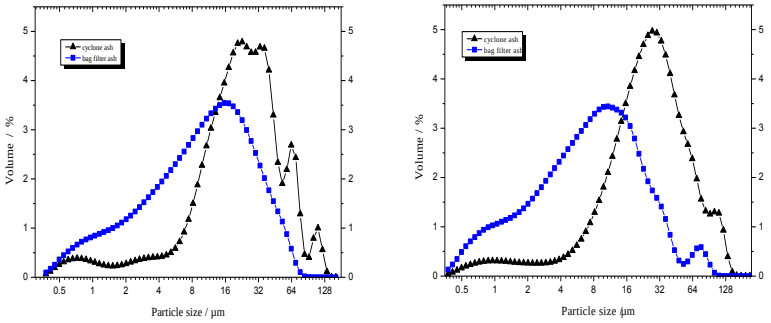
(b) bituminous coal

Figure 5 Relative enrichment factors of elements in different solid stream



(a) anthracite (b) bituminous coal

Figure 6 Particle size distribution of the IBHX solid and bottom ash



(a) anthracite (b) bituminous coal

Figure 7 Particle size distribution of the cyclone ash and bag filter ass

Highlights

> Pilot CFB combustor with multi-stage solid management. > Mass balance ratio ranges from 56%-137%. > Volatile elements tend to be enriched in fine particles. >Anthracite shows lower emission factors.



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Partitioning of trace elements, As, Ba, Cd, Cr, Cu, Mn and Pb, in a 2.5 MWth pilot-scale circulating fluidised bed combustor burning an anthracite and a bituminous coal

Duan, Lunbo; Sun, Haicheng; Jiang, Ying; Anthony, Edward J.; Zhao, Changsui

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Lunbo Duan, Haicheng Sun, Ying Jiang, Edward J. Anthony, Changsui Zhao, Partitioning of trace elements, As, Ba, Cd, Cr, Cu, Mn and Pb, in a 2.5 MWth pilot-scale circulating fluidised bed combustor burning an anthracite and a bituminous coal, Fuel Processing Technology, Volume 146, 1 June 2016, pp1-8

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