Effect of combustion temperature on the emission of trace elements under O_2/CO_2 atmosphere during coal combustion

Chengrui Qu[1], Mo Zhang[1], Michael. D. Mann[2]

[1]School of Energy and Power Engineering, Northeast Electric Power University, Jilin 132012, Jilin Province, China
[2]Institute for Energy Studies, University of North Dakota, Grand Forks 58202, North Dakota, USA

qcr88@163.com

Abstract. The effect of combustion temperature on the emission of trace elementswas studied under O_2/CO_2 atmosphere during coal combustion in a laboratory scale fluidized bed combustor. The elemental composition of fine fly ash particles collected with a low pressure impactor(LPI)was quantified by X-Ray F1uorescence Spectrometer (XRF). The elemental composition of coal and bottom ash was quantified byinductively coupled plasma-atomic emission spectroscopy (ICP-AES). The results indicate that the contents of Mn, Zn, Cd and Cr in the fly ash increase with the rise of combustion temperature. It is found that the enrichment of Zn and Cd is greater in the submicrometer particles than the supermicrometer particles, but Mn and Cr do not enrich in the submicrometer particles. Mn, Zn, Cd and Cr display one peak around 0.1μm. The relative enrichment factor (R_{ij}) of four elements is in the order of Zn, Cd, Mn and Cr. Zn and Cd are mostly retained in fly ashwhileMn and Cr are retained in both the fly ash and bottom ash.

1. Introduction

In additional tothe major elements Si, Al, Ca and Fe, many coals contain trace elements such as Pb, Ni, Mn, Zn, Cd, Cr, and Hg that aretoxic trace elements [1-3].

When the coal is burned in the furnace, the trace metals are released from the fuel and transformedinto ash. This can result in high concentrations of trace metalsin certain ash fractions. Consequently, due to the high heavymetal concentrations, the ash cannot be circulated back tothe soil, i.e. disposed of in conventional landfills or used as cement replacement and other reuse applications unless the heavy metals can be separated into a smallash sidestream.Additionally, these elements can be emitted into atmosphere during coal combustion, posing a serious threat to human health and the environment.

The behaviour of heavymetals inthe combustion processeshas been studied fairly extensively in the recent years.However, the work has mainly concentrated on coal combustion under air [4-8].During coal combustion in air, the combustion temperature and the gas atmosphere have been found to strongly affectthe volatilization behaviour of the ash-forming constituents [9-12].

O_2/CO_2 combustion, using oxygen and recycled flue gas as a combustion medium, can achieve a CO_2 concentration of more than 95% in dry flue gas to enable an easy CO_2 recovery. It can also offer additional benefits of substantially reducing NO_x, and SO_2 emissions. Therefore, O_2/CO_2 combustion is considered as one of the most promising technologies for coal-fired power plants to control CO_2emissions, which has motivated numerous studies covering many scientific and engineering fundamental issues on the application of this technology.

However, to date a few investigations have been performed on the emission characteristics of trace elements in O_2/CO_2 combustion[13-15].
In this paper, the emission characteristics of trace elements in O_2/CO_2 atmosphere were experimentally investigated in a laboratory scale fluidized bed.

2. Experimental

2.1. Test materials and conditions

Xuzhou bituminous coal was used in this study. The samples of 1gwere sieved between 105 and 154μm. The properties of the pulverized coal and ash composition are summarized in Table 1 and Table 2, respectively. The O_2 and CO_2 mixture was used as the combustion medium in the experiments to simulate the O_2/CO_2 combustion. The volumetric mixture ratio of 21%O_2:79%CO_2 was employed. The combustion temperature was set at 750℃, 850℃, 950℃ and 1050℃ to study the effect of combustion temperature on the emission of trace element. Fine particles were collected by LPI. The mass of particles was less than 1 mg on each stage of LPI in order to avoid bounce. The collecting time of particles was less than 30 minutes.

Table 1. Proximate analysis and ultimate analysis of Xuzhou bituminous coal

Proximate analysis（air dry basis）/ wt, %				Ultimate analysis（air dry basis）/ wt, %				
M	A	V	FC	C	H	O	N	S
0.99	18.19	38.46	42.36	68.30	5.31	2.58	0.78	3.87

Table 2. Analysis of ash composition of Xuzhou bituminous coal（wt%）

SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	PbO	TiO_2	K_2O	Na_2O
52.59	32.78	4.69	2.68	0.46	0.67	13.99	1.41	0.51

2.2. Experimental devices and analysis of sample

Coal samples were burned in a laboratory scale fluidized bed as shown in Fig. 1. The experimental system includes a fluidized bed, cyclone, LPI and gas battles.The ash particles were isokinetically collected at the exit of the furnace using a probe, which was connected to a cyclone and an LPI. The cyclone was used to remove flyash particles with diametersmore than 10μm and the LPI was employed to segregate the fine particles by size. The LPI is composed of 13 stages having 50% aerodynamic cut-off diameters ranging from 0.023 to 9.314 μm. A separate LPI sampling procedure was performed using Teflon membranes as the substrates. The settled particles on each stage were subjected to elemental analysis using XRF. The trace elements in coal samples and bottom ash were analyzed by ICP-AES.

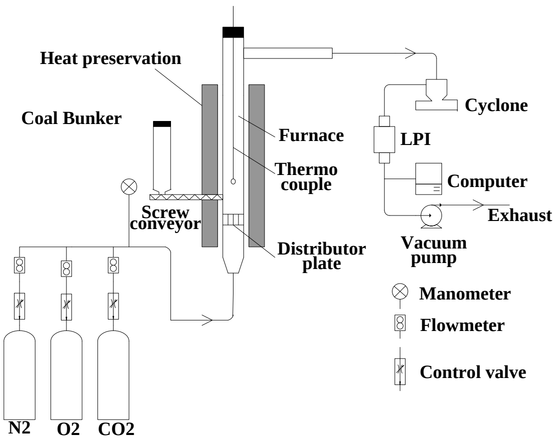


Figure 1. Schematic diagram of laboratory scale fluidized bed

3. Results and discussion

3.1 The effect of combustion temperature on emissions of trace elements

The effect of combustion temperature on emissions of Mn, Zn, Cd and Cr is presented in Fig. 2. The concentration ofthe four elements in the fly ash increaseswhen the temperature increases. Zn and Cd are present in the coal as sulfide mainlyand vaporize at relatively low temperatures.Mn and Cr are present in the coal as clay and due toit'smelting point, so they evaporate more difficultly when the coal is combusted.

The mechanism of trace element transformation during coal combustion is illustrated as follows: the vaporized metals at high temperature near the combustion flame will subsequently nucleate or condense at a lower temperaturedownstream. The conversion of vaporized components intovarious solid and/or liquid forms is the key factor influencing the final trace elements' transformation/partitioningbehaviour. It is determined, basically, by three complexand interrelated processes: adsorption, condensation andchemical transformation.

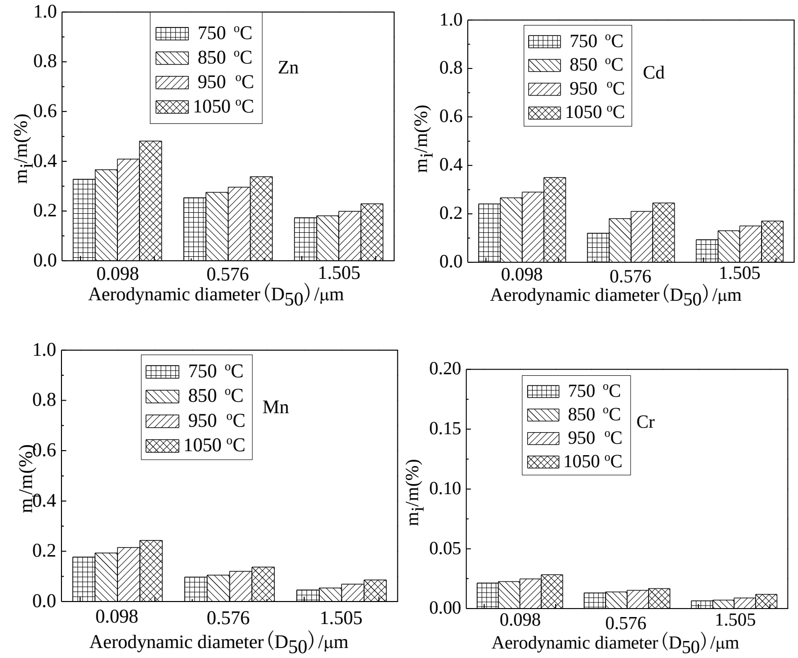
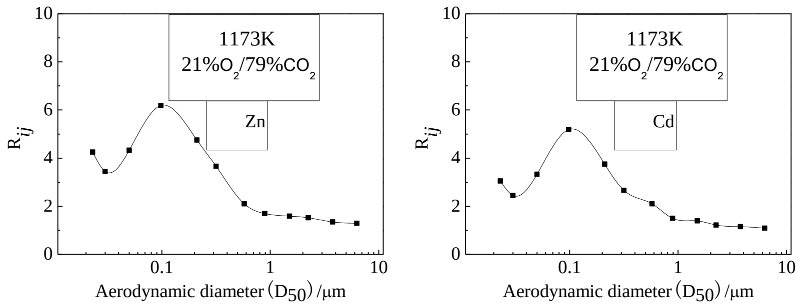


Figure 2. Effect of temperature emission of Mn, Zn, Cd and Cr

3.2The enrichments of trace elements

The enrichment of Mn, Zn, Cd and Cr in $PM_{2.5}$is shown in Fig. 3. The variation trends of elements in the PM are described by relative enrichment factorR_{ij}, that is, $R_{ij} = C_{ij}/C_{fj}$, where C_{ij} and C_{fj}stand for the mass fraction of element j in ith stage PM of LPI and bulk ash sample, respectively.

$R_{ij} = C_{ij} / Ci_{13}$ (1)



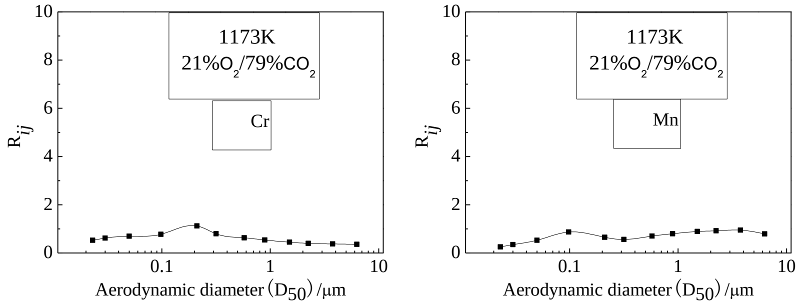


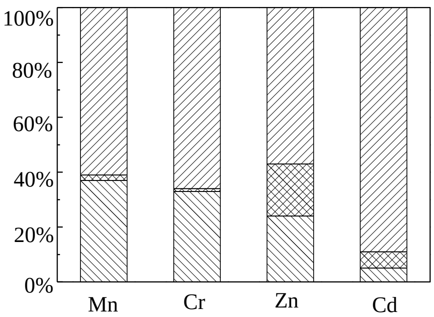
Figure3.distributionof trace elements in $PM_{2.5}$

The element enriches in particles of jth stage if R_{ij} is more than 1, otherwise, the element does not enrich. Zn and Cd enrich in PM_1 but Mn and Cr show little enrichment in PM_1. There is a peak in PM_1 around 0.1 μm. This is related to nucleation and condensation mechanism. When the coal burns, vapours of elements form very fine particles through homogeneous nucleation.These fine particles collide with each other and form PM_1. The nucleation and condensation mechanism can explain the phenomenon that volatile elements such as Zn and Cd enrich in PM_1.

3.3The partition of trace elements

Fig. 4 shows the partition percentages of Mn, Zn, Cd and Crin the fly ash, flue gas and bottom ash. The trace element partitioningis often referred to the dispersion of elements among different emission streams: bottom ash, fly ash and flue gas. We calculate itby a mass balance method from the following equation:$M_{ic} = M_{if} + M_{ib} + M_{ia}$ (2)

M_{ic}, M_{if}, $M_{ib}$and M_{ia} representthe element i content in coal, fly ash, bottom ash and fluegas,respectively.Fig. 4 illustrates that the mass percent of Zn and Cd in flue gas are greater than Mn and Cr. These trace elements are completely vaporized atcombustion temperature, and are sufficiently volatile toremain partly in the vapour phase as a very high vapour pressure at typical stack outlet temperature. Zn and Cd are mostly retained in fly ash and partly in bottom ash. Mn and Cr are mostly retained in fly ash and bottom ash.



（: fly ash； : flue gas； : bottom ash）

Figure 4. Partition of trace elements between bottom ash, fly ash and flue gas

4. Conclusions

Combustion of Xuzhou bituminous coal was conducted in a laboratory scale fluidized bed to investigate the effect of combustion temperature on emissions of Mn, Zn, Cd and Cr. The major conclusions are drawn asfollows:

(1) The content of Zn, Mn, Cd and Crin the fly ash increasewith increasing temperature. The extent of increase of four elements is more remarkable in PM_1 than PM_{10}.

(2)Enrichments of Zn and Cd are observed in PM_1, Mn and Cr do not enrich in PM_1.

(3) For the fixed particle size, the enrichment of the four elements is highest for Zn followed by Cd, Mn and Cr.

(4) Zn and Cd are mostly retained in fly ash and partly in bottom ash. Mn and Cr are mostly retained most heavily in the fly ash, but also have significant concentrations in the bottom ash.

5. References

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