



Research article

Behavior of selenium in the flue gas of pulverized coal combustion system: Influence of kind of coal and combustion conditions



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ARTICLE INFO

Article history:

Received 30 March 2017

Received in revised form 5 July 2017

Accepted 15 July 2017

Keywords:

Pulverized coal combustion

Flue gas

Selenium

Distribution

Kind of coal

ABSTRACT

Five kinds of coals (mainly low Se levels) were pulverized and combusted in a turbulent furnace combustion test rig, which had a burner on the top and a gas cooling pipe with two sampling ports at the side near the bottom. A sampling train according to ISO 17211:2015 was attached to the ports to assess the distribution of Se in the gas and solid phase (coal ash) in the flue gas. The fraction of Se in the solid phase (R_{Se_s}) in the flue gas is given by $R_{Se_s} = Se_s / (Se_s + Se_g)$, where Se_s and Se_g are the concentrations of Se in the solid and gas phases, respectively. For all coals, R_{Se_s} increased with a decrease in the flue gas temperature from 300 °C to 100 °C, and the difference in R_{Se_s} by the temperature was greatly dependent on the kinds of feed coals. The effects of the contents of minerals in feed coals and the combustion conditions upon R_{Se_s} were studied. The partitioning of Se from the gas to solid phase in the flue gas was elucidated in terms of the chemical and physical adsorption of Se on coal ash.

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1. Introduction

The discharge of Se compounds into the environment can have negative impacts on human health. Although Se is an essential element for humans and other animals at low levels of uptake, a high level of uptake can cause serious damage to living organisms [1,2]. In several countries, Se is listed in national pollutant chemical inventories, such as the United Kingdom's Pollution Inventory, Australia's National Pollution Inventory, and the Pollution Release and Transfer Register of Japan. In the future, regulations related to the control of Se emissions into air will be formed in many countries.

Combustion of coal is a major anthropogenic emission source of volatile toxic trace elements such as Hg and Se. It has been reported that various hazardous trace elements are distributed in the by-products of coal combustion systems, such as coal fly ash, flue gas desulfurization wastewater, and exhaust gas, and these elements are discharged into the environment in both gas and solid states [3–5]. The behavior of Hg in coal combustion systems has been well studied. The chemical speciation of Hg in coal combustion flue gas as well as the distribution of Hg in the combustion systems have been reported [6–10]. Moreover, the partitioning of Hg from the gas to solid phase (coal ash) in the flue gas has been investigated [11–13].

Compared with Hg, studies on the behavior of Se in coal combustion systems are scarce. Senior et al. investigated the behavior of some trace

elements, including Se, in a coal combustion system by using a cascade impactor to collect particulate ashes [14,15]. They suggested that Se is almost completely vaporized in the flame zone to form SeO_2 , while Se still exists in the gas phase in low temperature zones, such as at the entrance to electrostatic precipitators (ESPs), although the partitioning of Se from the gas to solid phase (coal ash) occurs considerably. However, compared with other elements, the mass balance of Se in the low temperature zones is not very high. A similar experiment was carried out by Seams et al. who reported that the concentration of Se in coal ash was dependent on its particle size whereas the relationship between the concentration of Se and that of Ca or Fe in size-segregated ash samples was not clear [16,17].

There have been some reports on the partitioning of Se from the gas to solid phase in the flue gas of coal combustion based on model and simulation studies. Lopez-Anton et al. investigated the retention of Se on the surface of coal ash using reference materials such as activated carbon and limestone, and suggested that the Se retention may be influenced by the presence of carbonaceous particles and Ca compounds in the ash [18,19]. Senior et al. reported that the partitioning of Se from the gas to solid phase may be ascribed to the reaction of gaseous Se with Ca and Fe compounds in feed coal in the flame zone, whereas the coagulation of gaseous Se leads to physical adsorption on coal ash in the low temperature zone [20]. In addition, Raeva et al. reported that the presence of Ca in feed coal promotes the Se retention in the solid phase due to common acid-base chemistry in coal combustion [21].

Noda et al. examined the behavior of some trace elements, including Se, in the flue gas of coal combustion [22]. They used US Environmental Protection Agency (EPA) method 29 [23], which allows for the

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Table 1
Properties of feed coals.

	Unit	Feed coal				
		A	B	C	D	E
Inherent moisture	wt% (air dried)	3.3	4.8	3.1	2.4	7.2
Ash		12.3	13.0	12.2	15.7	3.9
Volatile matter		34.4	32.7	31.4	26.0	43.0
Fixed carbon		50.0	50.0	53.3	55.9	46.0
Ash composition	wt% of ash					
	SiO ₂	60.4	47.8	56.8	46.6	38.1
	Al ₂ O ₃	23.3	41.8	21.0	29.6	22.6
	Fe ₂ O ₃	4.28	3.84	6.51	5.95	18.5
	CaO	3.20	2.11	3.50	6.95	7.37
	MgO	1.06	0.39	1.47	0.96	2.35
	Na ₂ O	0.32	0.23	0.60	0.36	1.14
	K ₂ O	1.40	0.51	2.04	0.60	0.95
Se	mg/kg (dry basis)	0.40	2.55	0.27	0.49	0.05

determination of distribution of the elements in gas and solid phase in the flue gas. However, it has been noted that EPA and related methods are not particularly effective in terms of Se speciation in coal combustion systems [24,25]. The main reason for this is the difficulty in sampling of Se in the gas phase, especially in the low temperature zone, because of the reduction of SeO₂ to Se⁰ by SO₂, which is usually present in large quantities in the flue gas. The Se⁰ species tends to adhere to the tubing and vessel walls in the gas sampling train. To prevent the problem, a new official method for the sampling and determination of Se in the flue gas was recently issued (ISO 17211:2015) [26]. This is an improved method for EPA method 29 in terms of the addition of a rinse process to attain the complete recovery of Se adhered to the tubing and vessel walls. Thus, the sampling of Se in the gas phase in the presence of SO₂ can be effectively carried out.

The Se contamination in desulfurization wastewaters has been one of the big problems in Japanese coal fired power plants. Therefore, the use of coals with low Se levels is currently recommended in Japan. In this study, the ISO method was applied to the combustion of various coals, including low Se level coals, in a turbulent furnace combustion test rig to

Table 2
Combustion conditions.

	Unit	Condition 1	Condition 2	Condition 3
Primary air	Nm ³ /h	6	6	6
Secondary air		52	36	36
Over-fire air		0	16	16
Over-fire air port (distance from the furnace top)	cm	–	116	166

assess the distribution of Se in the gas and solid phase (coal ash) in the flue gas. The influence of flue gas temperature, kind of coal, and combustion conditions on the behavior of Se was investigated in terms of the partitioning of Se from the gas to solid phase in the low temperature zone, which is modeled for NO_x removal systems (ca. 300 °C) and ESPs (ca. 100 °C).

2. Experimental

2.1. Coal samples

Five coals (termed A–E), which had been imported for use in Japanese coal-fired power plants, were tested. Analysis of these coals was conducted based on JIS M8812 [27] and the results are shown in Table 1. The concentration of Se in the coals varied from 2.55 to 0.05 mg/kg. The ash composition was obtained by X-ray fluorescence analysis (Zetium, PANalytical V. B.) after low temperature ashing of the coal. The determination of Se in the coal was conducted as follows. The digestion of coal sample was carried out using a microwave processor (ETHOS 1, Milestone Inc.) with the acidic mixture of HNO₃/H₂O₂. Detailed conditions of the microwave processing were described in our previous study [28]. The concentration of Se in the resulting solution was analyzed by hydrate generation atomic absorption spectrometry (HG-AAS; HYD-20 hydride generator, Nippon Jarrell-Ash Co. Ltd. and solar S2 spectrometer, Thermo Scientific Inc.) [29].

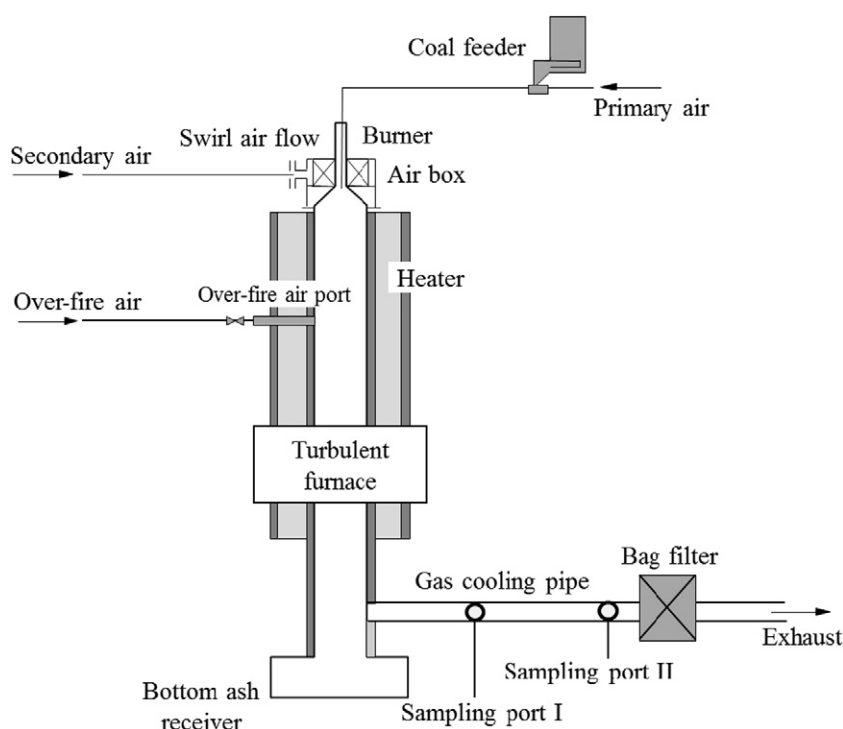


Fig. 1. Schematic of the turbulent furnace combustion test rig with sampling ports I and II to which the Se sampling train (Fig. 2) was attached.

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