



Mercury emission characteristics from coal combustion by supplying oxygen and carbon dioxide with limestone injection



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ABSTRACT

Coal combustion experiments at a lab-scale furnace with a capacity of 10,000 kcal/h were conducted in order to investigate the speciation changes and mass distribution of mercury with limestone injection in conditions of air and oxy-fuel combustion. Mass distribution and concentration of mercury in flue gas at oxy-fuel combustion were higher than those at air combustion. With limestone injection, the reduction of mercury from flue gas by adsorption into limestone could be clearly observed at both air and oxy-fuel combustion conditions. In the speciation of mercury, the particulate mercury in flue-gas was dominant with around 50% and the portion of elemental mercury was higher than that of oxidized mercury as shown at other coal combustion cases. At oxy-fuel combustion, most of particulate mercury seemed to be bounded to limestone more easily in comparison to air combustion, so that the elemental mercury in flue gas resulted to be more dominant at oxy-fuel combustion.

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1. Introduction

In oxy-fuel combustion which fuel is combusted at O₂ and CO₂ supplying condition, the emission trend of pollutants in the flue gas should be different than in the conventional air combustion. In the conventional air combustion, the flue-gas from the coal-fired power plants includes various air pollutants such as fine particulate, toxic trace metals, mercury compounds, sulfur oxides, and nitrous oxides. Linak et al. [1–3] have reported several studies on the chemical properties of the fine particulates formed in the coal combustion process. According to Linak et al., fine particulate are thought to originate from metal vapor at high combustion temperatures and they are formed via nucleation, condensation, and coagulation processes. The coal combustion resulted in fine particulate emissions which were primarily enriched by trace metals such as mercury. The temperature of the furnace and the residence time were the controlling factors and the particle size distribution depended on the interaction between the chemical reactions, nucleation, condensation, and coagulation [4]. Coal power plants release fine particulate enriched by trace metals such as mercury due to the limited removal efficiency

of the existing air pollutants control devices for these fine particulates [5]. Mercury can exist as elemental mercury (Hg⁰), oxidized mercury (Hg²⁺) and particulate mercury (Hg_p). Once in the air, mercury can be widely dispersed and transported thousands of miles from the emission sources. Mercury undergoes a series of complex chemical and physical transformations as it cycles among the atmosphere, land, and water [6, 7]. Lee et al. [8] investigated the speciation and mass distribution of mercury at bituminous coal-fired power plant. The facility was equipped with an electrostatic precipitator (ESP) and a wet flue gas desulfurization (FGD) as air pollution control devices (APCD). According to this study, elemental mercury was dominant at the outlet of FGD because oxidized mercury was primarily absorbed in the lime/limestone solution due to the solubility of oxidized mercury in water. Pudasainee and Kim et al. [9–12] compared the variation of mercury emission and speciation in APCD at various coal power plants in Korea. Those plants were equipped with selective catalytic reactor (SCR), ESP, and FGD and the configuration was different at each coal power plant. The removal efficiency of mercury was different at each coal power plant and depends on the configuration of APCD. The removal of mercury increased at the facility equipped with SCR, ESP, and wet FGD because the oxidation of elemental mercury by SCR enhanced the removal of oxidized mercury in the downstream of wet FGD. Researchers have proposed the use of several sorbent materials such as limestone for metal control. Owens et al. [13] performed

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equilibrium calculations for a number of toxic metal species reacting with sorbents in order to establish the relative efficiencies. For a specific sorbent and toxic metal, there is a temperature range over which it is most effective. Wu et al. [14] extended their earlier studies to the development of a multifunctional sorbent for the capture of sulfur dioxide, alkali metals, and lead. This aluminosilicate sorbent was effective in the simultaneous removal of the species and no loss of activity was observed due to competitive reactions. Zheng et al. [15] concluded that the coal oxy-fuel combustion had an effect on the amount and type of trace metals such as mercury emissions in the vapor phase. These fine particulate and trace metals such as mercury from oxy-fuel combustion would be difficult to capture by the APCD of the conventional coal power plant. Therefore, it would be necessary to figure out the emission trend of fine particulate and trace metals such as mercury from the oxy-fuel combustion. In this study, the speciation of the mercury compound was predicted both in the conditions of conventional air combustion and in the conditions of oxy-fuel combustion by a chemical equilibrium and applications (CEA) simulation code. Also, a lab-scale furnace experiment was carried out in order to compare the emission characteristics of mercury in the conditions of oxy-fuel combustion with that of conventional air combustion with limestone injection. The mass distribution and speciation of mercury in the flue-gas, fly ash and bottom ash were investigated in order to compare the transformation characteristics of mercury in the conditions of oxy-fuel combustion with those of air combustion with limestone injection.

2. Experimental methods

The experiments were conducted in a 10,000 kcal/h oxy-fuel combustion reactor, which based on the low-heating value of coal-fuel. Fig. 1 shows the schematic diagram of the lab-scale consisting of an air and gas supply, a coal feeding, a control box, two combustion chambers, a heat exchanger and a bag filter. The air and oxy-fuel combustion experiments were conducted by using the bituminous coal as fuel. The basic characteristics of the coal fuel are presented in Table 1. With coal elemental analysis, CEA code simulation was conducted in order to predict the speciation of mercury such as elemental mercury (Hg^0) and oxidized mercury (Hg^{2+}) in the flue gas from air and oxy-fuel

Table 1
Basic characteristics of coal fuel in lab-scale experiment.

Low heating value, kcal/kg		5966			
Proximate analysis (%)	Volatile	35.5	Major element composition (wt.%)	Al	1.113
	Fixed carbon	56.7		K	0.085
	Ash	7.7		Ca	0.861
Elemental analysis (%)	C	76.5	Ni	0.002	
	H	4.6	Zn	0.001	
	N	0.2	Ni	0.002	
	S	0.17–0.3	Hg	0.046E–04	
	O	18.4	Cl	0.7	

combustion. Comparative studies of coal combustion in air and oxy-fuel combustion were carried out with the stoichiometric ratio of 1.265 and 1.165, respectively. The limestone injection to the furnace was conducted in the conditions of air and oxy-fuel combustion for the desulfurization test. Sampling points of gaseous, bottom ash and fly ash were presented in Fig. 1. The combustion temperature in the lab-scale furnace was approximately 900 °C. The experimental conditions are summarized in Table 2. The mercury sampling in the flue-gas and the analysis were conducted in the outlet of the combustor by the Ontario Hydro Method ASTM D6784 [16]. The Ontario Hydro Method was developed in order to measure elemental mercury, oxidized mercury, particulate mercury, and total mercury from combustion sources. Solid samples were pre-treated by US EPA Method 7470A [17]. A cold vapor atomic absorption spectroscopy (CVAAs) mercury analyzer (RA-915+, LUMEX) was used to determine the mercury speciation in the flue-gas and the mass distribution in the gaseous and solid samples such as the bottom ash and fly ash.

3. Results and discussion

3.1. Desulfurization process and mercury transformation

The essential problem with flue gas desulphurization (FGD) from coal combustion is its inefficiency. Limestone is typically used to capture

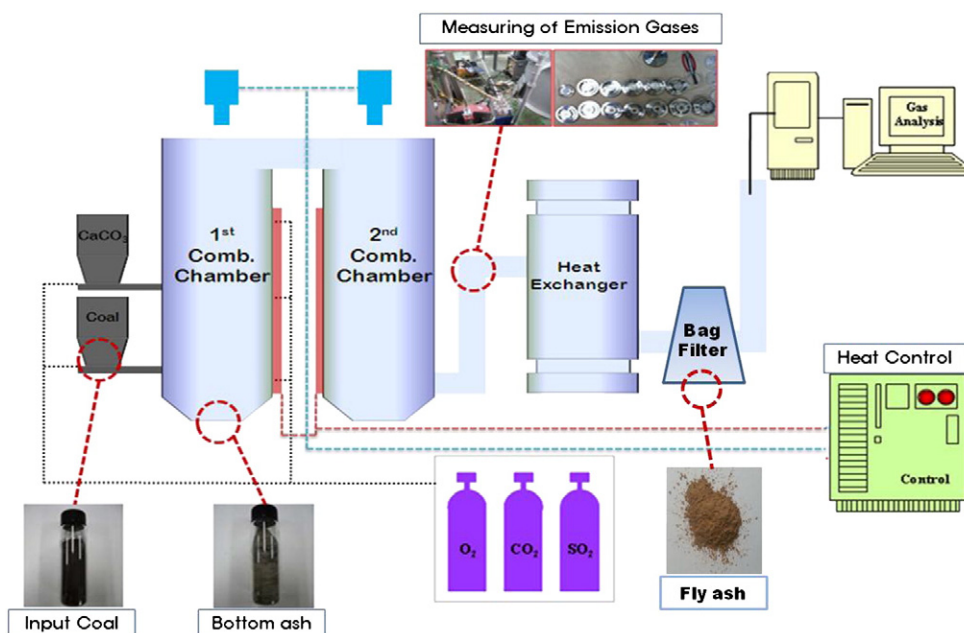


Fig. 1. Schematic diagram of experimental oxy-fuel combustion facility.

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