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Mercury emission and speciation in fly ash from a 35 MW_{th} large pilot boiler of oxyfuel combustion with different flue gas recycle

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HIGHLIGHTS

- The distribution of Hg in a 35 MW_{th} large pilot boiler of oxyfuel combustion was investigated.
- Higher concentration of gaseous mercury is contained in oxyfuel combustion flue gas.
- The oxyfuel combustion atmosphere enhances the conversion of Hg^0 to Hg^p.
- Trigonal red HgS is the main mercury species in air combustion fly ash.
- Mercury bound to organic matter (Hg–OM) and trigonal red HgS are the main species in the oxyfuel combustion fly ash.

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ABSTRACT

The behaviour of mercury is investigated in a 35 MW_{th} large pilot boiler of oxyfuel combustion with dry and wet flue gas recycle (FGR). The mercury species in fly ash particles as well as the involved reaction mechanism are systematically investigated. The results suggest that higher concentration of gas–phase mercury (Hg^g) is contained in oxyfuel combustion flue gas. The conversion of Hg^g to particulate associated mercury (Hg^p) is enhanced in oxyfuel combustion. Different mercury species are presented in fly ash collected from air and oxyfuel combustion: trigonal red HgS is the main mercury species in air combustion; mercury bound to organic matter (Hg–OM) and trigonal red HgS are both dominant in the samples from oxyfuel combustion. The prominent difference is the significantly increase of Hg–OM in oxyfuel combustion atmosphere. The oxyfuel combustion atmosphere would favor the formation of oxygen–rich functional groups, especially the C=O group, which is responsible for the adsorption of mercury and formation of Hg–OM in fly ash.

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1. Introduction

Emissions of CO_2 from coal–fired power plants is one of the major environmental concern. Oxyfuel combustion is one of the most promising technologies for capturing CO_2 from power plants [1]. The coal particles are burned with oxygen stream in the oxyfuel combustion system and a proportion of CO_2–rich flue gas is recycled to the boiler with an intention to improve the heat transfer. In the previous studies, much attention had been paid to the operational and environmental impacts, such as ignition, flame temperature and propagation, char combustion, heat transfer and pollutant formation [1–4].

The fate of mercury is important for the operation of oxyfuel combustion because of its environmental concern [5,6] and potential corrosion for CO_2 compression and purification units [7,8]. In the past few decades, extensive studies had been carried out to understand the fate of mercury in conventional air combustion [9–15]. As the most volatile trace elements in coal, most of the mercury is volatilized as gaseous elemental mercury (Hg^0) at high temperature combustion zone. Cooling the flue gas and interactions of Hg^0 with other flue gas composition could result in the oxidation of Hg^0 to oxidized mercury (Hg^{2+}). Meanwhile, several part of Hg^{2+} could be converted into particulate bound mercury (Hg^p) with the participation of fly ash and flue gas.

Thermodynamic modeling showed that Hg^0 is the primary species over a wide range of temperatures (800–1400 °C) in oxyfuel combustion, with a small portion of HgO (g) at low temperature

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[9]. The increase of chlorine content in combustion atmosphere, particularly at low temperature, could facilitate the formation of oxidized mercury (Hg^{2+}). A combined homogeneous–heterogeneous model developed by Gharebaghi et al. [16] showed that the mercury retention on fly ash particles depended on temperature, burnout and particle sizes. Meanwhile, a higher amount of particulate mercury was occurred in the oxyfuel combustion conditions. However, the species and fate of mercury during coal combustion is complex and it depends on many factors, such as the combustion temperature, the mass and heat transfer, the content of Hg and Cl/Br in the coal, the flue gas composition, the fly ash components (i.e. unburned carbon (UBC), Fe_2O_3, CaO) and the application of gas cleaning equipment (SCR, ESP/FF, FGD) [17,18]. Moreover, in the oxyfuel combustion unit, impurities in flue gas like SO_2, SO_3, HCl, and H_2O is much higher than those in air combustion because of the flue gas recirculation [19–21]. This could result in the difference of the species and fate of mercury between oxyfuel and air combustion.

Recently, several results about the fate of mercury under oxyfuel combustion conditions have been reported over pilot–scale measurements. Font et al. [22] found that, in the exhaust gas from a 90 kW_{th} bubbling fluidized bed oxy–combustion pilot plant, about 82% of the gaseous Hg was occurred in Hg^0. Similar percentage of Hg^0/Hg^{2+} in the exhaust gas from a 10 kW_{th} fluidized bed under oxyfuel combustion condition was also obtained by Roy et al. [23], where the percentage of Hg^0 in the total gaseous mercury was about 83%. Zhuang et al. [24] investigated the fate of mercury during oxyfuel combustion in a 173.5 kW_{th} experimental system with different flue gas recycle. They found that the conversion of Hg(g) to Hg(p) and Hg^0 to Hg^{2+} were both promoted in oxyfuel combustion. However, different conclusions were obtained by the study of Obras–Loscertales et al. [25], where the recirculation of SO_2 inhibited the Hg^0 oxidation and thus the mercury fixation as particle–bound mercury decreased. Up to date, the data are mostly based on bench and pilot–scale measurements, while information from industrial–scale facility are limited. Córdoba et al. [10] reported the first study of Hg species and behaviour in a 20 MW_{th} industrial–scale oxyfuel combustion plant. The results revealed that the continuous CO_2–rich flue gas recirculation played a significant role on Hg partitioning and fate. Even so, the data originated from industrial–scale oxyfuel combustion power plants is still very limited, especially the behaviour of mercury in oxyfuel combustion with different flue gas recycle (FGR). Moreover, the reaction mechanism between mercury, flue gas components and fly ash particles under oxyfuel combustion conditions are not clear as well.

To address these gaps, the emission and speciation of mercury in fly ash was investigated in a 35 MW_{th} large pilot boiler of oxyfuel combustion (HUST, Yingcheng, China) with dry and wet flue gas recycle (FGR). The mercury species in fly ash particles were investigated by a technique of mercury temperature programmed desorption (Hg–TPD) combined with sequential extration. The possible reaction mechanism between mercury, flue gas components and fly ash particles were analyzed as well. Such systematically investigation is of fundamental importance in solving environmental concern of mercury in industrial–scale oxyfuel combustion power plants.

2. Experimental facility and test procedure

The experiments were performed in a 35 MW_{th} large pilot boiler of oxyfuel combustion (HUST, Yingcheng, China), which was designed to operate with conventional air and oxyfuel combustion conditions (Fig. 1). During the oxyfuel combustion, pure oxygen mixed with recycled flue gas was feed into combustor. In this work, dry and wet FGR were carried out, respectively. For comparison, the test of air combustion was also performed at the same coal feeding rate as that of oxyfuel combustion. The properties of the feed coal used in the experiments are shown in Table S1 (Supplementary content).

The distribution of Hg_g and Hg^p in flue gas were both determined under air and different oxyfuel combustion conditions. Hg_g was measured at the ESP inlet, ESP outlet, and FGD outlet by EPA 30B method. Traps (Ohio Lumex 30B Carbon Traps) filled with activated carbon were used to collect the total mercury ($Hg^T = Hg^0 + Hg^{2+}$). In each test, two parallel traps were used, which was maintained at 120 °C. A constant gas flow rate of 35 mL min^{-1} was maintained for 30 min for each trap. Hg^p in fly ash particles were collected from the air preheater outlet by impactor and from ESP inlet by aspirated isokinetic sampler, respectively. For comparison, the fly ash particles at the right and left of air preheater outlet were collected at the same time. After sampling, Hg content in traps and fly ash were analyzed by RA–915M mercury analyzer with PYRO–915+ pyrolysis equipment (Ohio Lumex Company), where the detection limit (dl = 3σ) is 2–5 ppb. To minimize the experimental error which is missing in the operating conditions, the sampling was performed when the operating condition was stable for at least 1 h and each test repeated 3 times.

The identification of mercury species in fly ash might contribute to understanding the mercury behaviour in combustion processes, and mercury temperature programmed desorption (Hg–TPD) is an

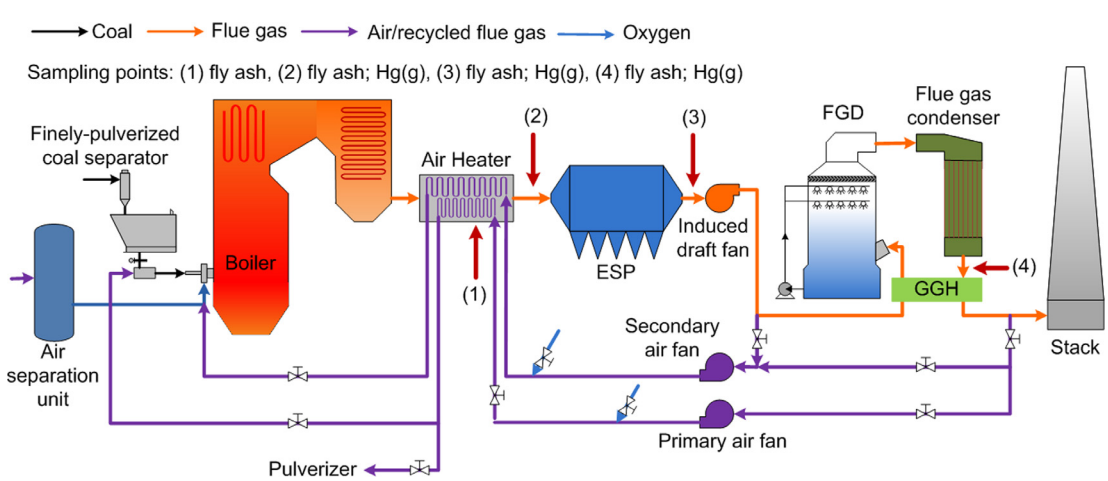

Fig. 1. Experimental schematic diagram and sampling points.

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