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Speciation and capture performance of mercury by a hybrid filter in a coal-fired power plant

Jin-Ho Sung^a, Seung-Ki Back^a, Bup-Mook Jung^a, Youn-Suk Kang^b, Chul-Gyu Lee^b,
Ha-Na Jang^{a,*}, Yong-Chil Seo^{a,*}

^a Dept. of Environmental Engineering, Yonsei University, Wonju 26493, Republic of Korea

^b J-E Tech Co., Ltd., Seoul 08584, Republic of Korea

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ABSTRACT

Mercury speciation and removal were investigated for a newly developed hybrid filter (HF) originally designed to remove fine particulates and retrofitted to the exhaust cleaning system in a coal-fired power plant. The test plant used a blend of anthracite and semi-anthracite coals for combustion. At the boiler outlet, Hg⁰ was the dominant mercury species in the flue gas at 89% of total Hg. After passing through the electrostatic precipitator (ESP) and HF, Hg²⁺ became the dominant species owing to oxidation of Hg at the lower temperature and interaction with other air pollutants in the flue gas. The Hg²⁺ subsequently accumulated as Hg_p by bonding with fly ash. Due to higher oxidation of Hg in the HF than in the ESP, more Hg²⁺ could be removed by the flue gas desulfurization (FGD) system through dissolution of Hg²⁺ when an HF was included. By installing hybrid filters alongside the existing devices, removal efficiencies of 86.7% and 99.3% for Hg and fine particulates, respectively, could be achieved, as opposed to 62.4% and 95.2% without the installation. Applying HFs in the existing air pollution control device (APCD) configuration was therefore recommended as the best simultaneous control technology for Hg and fine particulate matter (PM) at the test plant.

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1. Introduction

Mercury (Hg) compounds, present in trace amounts in the environment, have an adverse effect on human health owing to long-term exposure, contamination, and bioaccumulation (Gardner et al., 2010; Grandjean and Budtz-Jørgensen, 2007; Sarabia et al., 1998). They can also be transported long distances by the wind. Facilities with Hg emissions can be classified into coal-fired power plants and industrial boilers, incineration facilities, cement clinker production plants, and non-ferrous metal smelters. While flue gas from coal-fired power plants has low concentrations of Hg in the emitted gases, the total emitted volume is very large (Ito et al., 2006; Romanov et al., 2012). Consequently, flue gas is classified as a major source of Hg-emissions. Hg-emissions depend on the coal type, combustion technologies, and configuration of air pollution control devices (APCDs) used in a plant (Galbreath and Zygarrlicke, 2000). The APCDs that have been installed to date are primarily for the control of sulfur dioxide (SO₂), nitrogen oxides (NO_x), or particulate matter (PM) from the flue gas.

In coal-fired power plants, mercury is volatilized and converted into gas-phase elemental mercury (Hg⁰) at high temperatures. Elemental

mercury (Hg⁰) is converted to oxidized mercury (Hg²⁺) in the gas phase and particulate mercury (Hg_p) as a condensate on solids through a series of reactions in the APCDs. Mercury therefore exists in the flue gas as elemental mercury (Hg⁰), oxidized mercury (Hg²⁺), and particulate mercury (Hg_p). Mercury can be controlled by the existing PM, sulfur dioxide, and nitrogen oxide APCDs, in a “co-benefit effect” (Clack, 2009; Scala and Clack, 2008). According to the mercury data from Information Collection Request (ICR), fabric filters used for PM control can be very effective in mercury removal. Electrostatic precipitators are not as effective in removing mercury as the fabric filter configurations in APCDs. At the outlet of a boiler, Hg in flue gas is dominantly present as Hg⁰, which is generally uncontrolled and emitted to the atmosphere (Lee et al., 2006). In general, Hg speciation changes while passing through APCDs as a result of improved oxidation parameters, such as the flue gas temperature, halogen species present, and gas composition (Wang et al., 2009). Hg²⁺ can be adsorbed on fly ash or captured by wet APCDs (Galbreath and Zygarrlicke, 2000). Although mercury is beneficially captured by various APCD configurations, flue gas emitted into the atmosphere still contains a high proportion of elemental mercury because of the number of complex reactions in the entire process.

In this study, a hybrid filter (HF) was installed in an operational coal-fired power plant, in which blended anthracite and semi-anthracite coals were burned. The HF, which integrates technology from an electrostatic precipitator (ESP) and a fabric filter (FF) in a single unit, and

* Corresponding authors at: Dept. of Environmental Engineering, Yonsei University, Wonju 220-710, Republic of Korea.

E-mail addresses: janghana@yonsei.ac.kr (H.-N. Jang), seoyc@yonsei.ac.kr (Y.-C. Seo).

which was originally investigated for the removal of fine particulates, was operated to investigate Hg speciation changes and removal. A chemical equilibrium and application (CEA) simulation was conducted to predict Hg speciation in the coal-fired power plant. Hg speciation and flue gas composition were investigated to analyze the effects of flue gas properties and APCD configuration on Hg removal. Based on the test results, the optimal APCD configuration was determined for achieving the best Hg removal efficiency at the test plant.

2. Experiment and method

2.1. CEA simulation code

The behavior of Hg compounds can be predicted using the CEA simulation code, which uses a thermodynamic matrix to minimize Gibbs and Helmholtz free energies. The CEA is a program which calculates chemical equilibrium product concentrations from any set of reactants, and determines thermodynamic and transport properties for the product mixture. Built-in applications include calculation of theoretical combustion properties, and the program has a thermodynamic properties database for about 2000 chemicals. The CEA is used extensively in the fields of aerodynamics and thermodynamics (Senior et al., 2000).

2.2. Test facility and sampling method

In the coal-fired power plant, blended anthracite (20%) and semi-anthracite (80%) coals were used as fuel for combustion. The basic properties of these coals are shown in Table 1.

The APCD configuration and locations of the sampling points in the 400 MW coal-fired power plant are shown in Fig. 1. The main stream of flue gas passed through the existing APCDs, which consisted of an ESP and a FGD system, while 20% of the flue gas was taken out to pass through an HF for dust removal and Hg testing. The capacity of the HF was 240,000 m³/h, and it was installed between the ESP and the FGD systems. At the inlet of the ESP, the flue gas temperature was around 450 °C. After passing through the ESP, the flue gas temperature decreased to less than 150 °C. With the reduced temperature at the ESP outlet, heat damage to the filter bags in HF could be prevented. Hg speciation, dust concentrations, and gas compositions were sampled by the Ontario Hydro Method, a semi-auto-type dust sampling toolkit, and a gas analyzer (KINSCO Technology, Hymeth Plus Model, Republic of Korea). For the Hg compounds, the adsorbing solutions were analyzed by cold vapor atomic absorption spectrometry (Ohio Lumax company, Inc., RA215+, Russia).

2.3. Hybrid filter

As shown Fig. 2, the HF has an ESP and FF combined in a single chamber for simplification of the structure and minimization of installation area. In the HF chamber, a bag filter was installed between the dust collecting plates in front of the discharged electrodes. The HF was

made up of 4 chambers with 120 bag filters. During the HF operation, 3 chambers were operated at same time and another chamber has cleaning time for maintaining pressure drop. PM capture by the HF goes through the following process: (1) combustion flue gas from the boiler is injected at the inlet of the HF; (2) fine PM is captured when the flue gas penetrates the dust collecting plate and discharge electrode; (3) once the PM enters the electrode field, it is negatively charged by high-energy electrons of the corona; (4) the PM is collected on oppositely charged plates; (5) excess PM in the flue gas is removed by the bag filters. Filter loading was reduced since over 90% of the charged dust was removed by the dust-collecting plate located between the filter bags. The dust-cake layer was removed by pulsed air and then collected on the plates. The dust cleaning cycle was set to perform when filter bags in HF reached a predetermined pressure. In this way, a pressure drop of less than 100 mm Aq was maintained for 1 year. Existing ESP has the disadvantage of momentary increments of dust-emission by rapping period. A major drawback of existing FFs is the damage that occurs to the filter bag owing to the pulsed jet of air. In contrast, the HF has a longer cleaning cycle than a FF, so that not only is it possible to reduce energy and maintenance costs, but removal of contaminants in flue gas by the dust-cake layer is also expected. One of the effective advantages is the ability to reduce fine particulates and gaseous pollutants, including mercury, simultaneously. Once the bulk of the PM was removed by the dust collecting plate in the HF, the PM flowing into the bag filter in the HF was much less than what would flow into an only FF. After the capture of PM on the plate, the pressure drop caused by the dust cake on the filter surface in the HF would be decreased. The removed PM, which was charged by an electrode corona in the HF, had a thicker porous than in the filter bags in the HF, causing a lower pressure drop for the HF. As the results indicate, the surface area of the dust cake would increase, thus increasing the co-capture efficiency of Hg and fine PM.

3. Results and discussion

3.1. Prediction of Hg behavior

Fig. 3 shows the CEA simulation results for the speciation behavior of gas-phase Hg with the given elemental composition (Table 1) of coal used in the coal-fired power plant. As shown in the figures, Hg²⁺ was converted to Hg⁰ at 700 K when the air ratio was 1.0. An excess air ratio of 1.2 was also simulated for the test facility. As the temperature increased to greater than 700 K at the boiler outlet, Hg speciation at this point was predicted to be dominated by Hg⁰. However, since the flue gas temperature decreased while passing through an APCD such as the ESP, most of the Hg⁰ was oxidized through complex catalytic reactions between the fly ash and air pollutants such as NO, SO₂, and Cl₂ (Pudasainee et al., 2012). The resulting Hg²⁺ adsorbed on the fly ash, where it could be controlled by the PM control devices (Lee et al., 2013), thus enhancing the capture efficiency of gas-phase Hg owing to a co-benefit effect of the existing APCDs. Based on these results, it was likely that Hg was oxidized while passing through the HF, and Hg²⁺ bonded to the PM by catalytic reactions in this temperature range. After that, Hg_p could be captured by the HF.

3.2. Hg speciation and flue gas composition

The concentrations of SO₂, NO, and Hg species in the APCDs are shown in Table 2. Hg⁰ transformation affected its removal efficiency. Hg⁰ oxidation led to an increase in total Hg removal efficiency in the wet-type APCDs. Also, Hg²⁺ was converted to the solid-phase Hg_p. Amounts of total Hg captured by adsorption of fly ash in the APCDs increased with decreases in temperature and the amount of carbon in fly ash (Hower et al., 2010). As a result, total Hg concentration gradually decreased as flue gas passed through the APCDs. In the test facility, total Hg concentrations at the boiler outlet and stack were reduced from 7.43

Table 1
Basic properties of coals.

Method	Coal type combusted		Unit
	Anthracite	Semi-anthracite	
Calorific value analysis	5209	6469	kcal/kg (dry basis)
Proximate analysis	Moisture	2.74	2.15
	Volatile	5.44	16.76
	Fixed carbon	58.27	60.61
	Ash	33.55	20.48
Elemental analysis	Carbon	62.79	71.33
	Sulfur	0.58	0.44
	Chlorine	0.15	0.15
	Mercury	44.96	318.51

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