



Full Length Article

Mercury removal and its fate in oxidant enhanced wet flue gas desulphurization slurry



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ABSTRACT

Mercury is a toxic heavy metal that, once emitted or released, persists in the environment and circulates between air, water, sediments, soil and living creatures. Therefore, international governments and other authorities are taking measures to control mercury emissions from various sources. Despite many efforts, mercury remains a problematic pollutant in coal-fired installations in regards to differentiation of existing forms and their behavior in flue gas stream and purification units. Scientists try to understand its behavior in the flue gas and to capture it in one place, employing processes of adsorption, absorption, membranes or different catalysis. At the same time, researchers are also developing efficient and economically feasible technologies for mercury control. One such technology involves the capture of mercury in flue gases via gas-cleaning units through co-benefit application. Examples include, for instance, carbon injection in ESP, catalytic conversion in SCR unit, and absorption in a wet desulfurization scrubber.

This paper outlines a mercury capture method developed in American and Polish laboratories and will present the pilot-scale research with emphasize on the mercury behavior in the slurry with and without any added reagents.

1. Introduction

Environmental pollution, which highly influences the quality of human life, is nowadays a problem all around the world. One of the biggest sources of pollution comes from the combustion of coal, which can potentially liberate huge amounts of SO₂, NO_x, particulates, and heavy metals (such as mercury, arsenic and chromium) to the atmosphere. All of those pollutants are damaging to human health and to the environment. Coal can be used in a variety of industrial process, but is most commonly used in electricity and heat production at coal-fired power plants. The largest amounts of coal consumption in the power sector take places in the USA, China, Russia and India. Poland, where more than 83% of electricity production comes from conventional power plants, is one of the biggest coal consumers in Europe [1]. All power units in Poland are equipped with some air pollution control devices, especially electrostatic precipitators (ESPs) and flue gas desulphurization (FGD) scrubbers. Nowadays they are modernized or exchanged, because of the EUs Directives, which requires reducing SO₂, NO_x and particulate matter (PM) two-to-three times compared to

existing levels. New technologies for controlling NO_x emission, mainly selective catalytic reduction (SCR), are installed in Polish power plants too.

Nevertheless, these solutions do not sufficiently protect against emission of heavy metals to the atmosphere. Fossil fuel combustion is responsible for 24% of global anthropogenic emissions of mercury, which is highly toxic, especially to the nervous system of developing fetuses and young children [2]. This the reason why many international governments, agencies, and researchers have worked together to take actions to reduce mercury emissions from various sources [2,3]. Technologies that show promise to control mercury emissions include activated carbon injection (ACI) and co-benefit application of an SCR unit and a wet FGD scrubber [4–11]. Due to the competition between mercury and sulfur trioxide (SO₃) for active sites, ACI is most appropriate for low sulfur containing coals and is in widespread commercial use in the USA [12]. The ACI option can, however, increase operation costs because the presence of activated carbon in the fly ash can render it less attractive as a construction material (e.g., as concrete admixture).

For co-benefit technologies utilizing existing wet scrubber pollutant

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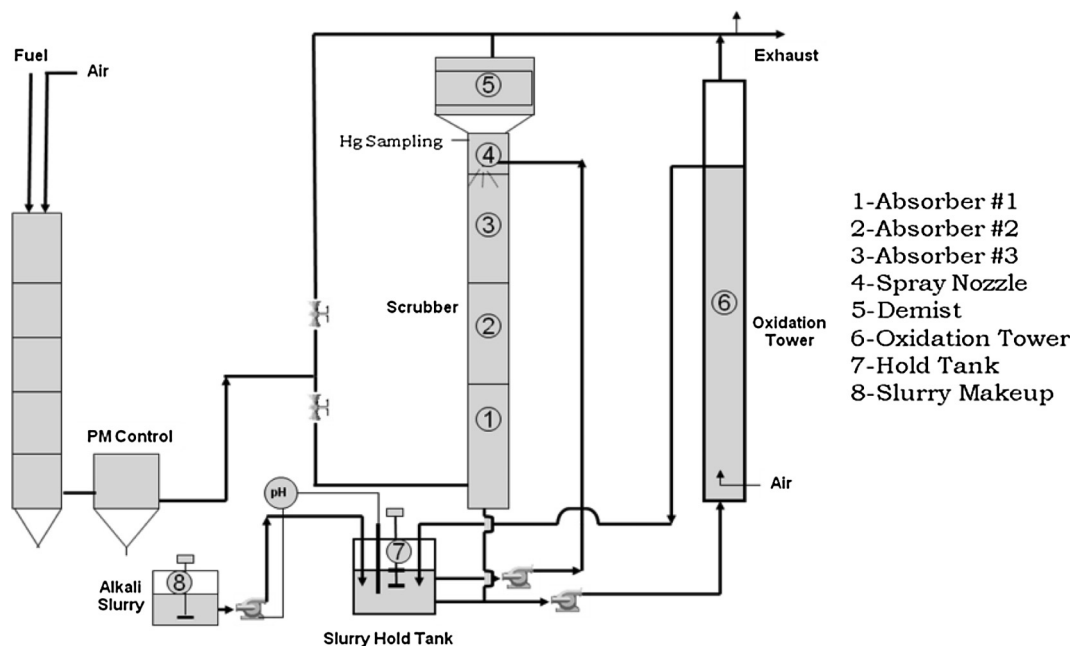


Fig. 1. Schematic of the pilot-scale wet scrubber system.

control devices, the presence of an SCR unit (used for NO_x control) is likely necessary to make this option attractive [5,6,10,11]. Because wet flue gas desulfurization (WFGD) systems are already installed at many coal-fired power plants, utilizing those scrubbers to also capture mercury seems an obvious and rational choice. However, WFGD cannot directly scrub elemental mercury vapor (Hg^0) efficiently, so it is necessary to find a cost-effective method to convert the Hg^0 to an oxidized or even a particle-bound form before entering the WFGD system. Conversion of Hg^0 to Hg^{2+} or particulate-bound mercury can be accomplished by heterogeneous catalysis or homogeneous gas phase oxidation. Injection of an oxidant into the flue gas (upstream of WFGD) to oxidize Hg^0 appears to be the simplest method of implementation. The biggest challenge is the selection of the proper oxidant and identification of the optimum process parameters [5,13]. Zhao and Rochelle [14] have studied Hg^0 absorption in aqueous sodium hypochlorite (NaClO). They found that Hg^0 can be significantly captured in such an oxidizing solution even at unfavorably high pH. Potassium hypochlorite (KClO) was also studied as an oxidizing reagent by Liu et al. [15]. In addition to providing some conclusions on the impact of slurry pH, their results indicated that Hg removal is favored under higher concentrations of KClO . Different chlorine-containing oxidizing agents were also screened with bench-scale tests by authors of this paper [13,16]. Of all examined agents, sodium chlorite (NaClO_2) was determined to be the most effective. With 8 mM (Mm) NaClO_2 in presence, it was discovered that the FGD CaCO_3 slurry could oxidize and capture nearly 100% of the mercury and 30–50% of the NO from a simulated coal combustion gas stream containing 200 ppmv NO_x , 1500 ppmv SO_2 and 206 $\mu\text{g}/\text{m}^3$ Hg^0 [13].

Significant amounts of NO removal by wet scrubber with oxidizing reagent were also demonstrated by other researchers [17–19]. In addition, to Cl-based species, potassium persulfate ($\text{K}_2\text{S}_2\text{O}_8$, KPS) catalyzed by Ag^+ and Cu^{2+} was also studied by Xu et al. [20] in a glass bubble column reactor. In the presence of 5.0 mM KPS, their mercury removal efficiency reached 75.4% and 97.0% for an Ag^+ concentration of 0.1 mM and 0.3 mM, respectively. All of these results demonstrated that a WFGD scrubber could be utilized as a co-benefit multipollutant control device if an SCR or an electrostatic precipitator is not installed in the plant.

Although oxidizing agent injection shows promise for Hg control, the current available data are limited to bench-scale studies. The

technical and economic feasibility must be assessed at pilot-scale prior to usage at large-scale in all of the discussed cases. However, before beginning large-scale research, it is necessary to more fully investigate and understand the chemistry of the process. For instance, when burning low halogen-containing coals, supplemental Cl or Br species may be required to oxidize Hg^0 and remove to acceptable levels. According to some research, Br is more effective than Cl in converting Hg^0 into its oxidized form [5]. Many chemical kinetic models and laboratory studies have indicated that chlorine increases the percentage of oxidized Hg in hot flue gas ($> 500^\circ\text{C}$), however the reaction between Cl_2 and Hg^0 has been shown to be too slow to be effective at lower temperature ($> 300^\circ\text{C}$) [21]. It is worth noting that all of the above mentioned research took into account mainly chlorine (Cl_2) species. Chlorite (ClO_2^-) species, which are investigated by the Authors of this paper, were not taken into account in those analyses.

Elemental mercury existing in the flue gas is oxidized to a form which can be subsequently captured in the slurry due to its high water solubility [5,13–15]. Adding an oxidizing reagent is also beneficial for NO removal because of the transformation of NO into more water soluble NO_2 , NO_2^- or NO_3^- species [17–19]. Authors have previously reported the bench-scale results [13,16] using NaClO_2 with a CaCO_3 slurry to simultaneously control SO_2 , NO_x and Hg emissions. This paper continues to validate its application at a pilot scale, with a focus on mercury removal from flue gas, discovering the fate of the captured mercury, with and without reagents, in wet scrubber and identifying the role which SO_2 and $\text{SO}_3^{2-}/\text{HSO}_3^-$ plays in the mercury capture process in a pilot-scale installation.

2. Experimental section

Research was performed in a pilot-scale test facility shown in Fig. 1. The system included a spray tower and a forced oxidation tower. The spray tower consisted of three absorbers (marked ①, ② and ③ in Fig. 1). Each absorber was 10 cm in diameter, 92 cm in length, and contained a 20-cm deep bed of plastic hollow balls (2 cm in diameter). The hollow balls were supported by a grid at the bottom of each absorber and fluidized by the upward flow of flue gas. Slurry was not discharged to the slurry hold tank (marked ⑦ in Fig. 1), but to a discrete 150-L oxidation tower (marked ⑥ in Fig. 1). The hold tank contained ~10 L of 5 wt% calcium sulfate (CaSO_4) for pH adjustment purposes. As shown

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