



Reduction of elemental mercury in coal-fired boiler flue gas with computational intelligence approach

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ABSTRACT

Mercury is an important pollutant emitted from coal-fired power plants. Elemental mercury (Hg^0) is harder to be removed than oxidized mercury (Hg^{2+}) and particulate bound mercury (Hg_p) in the flue gas at back-end of furnace. In this study, a method based on computational intelligence was proposed to enhance Hg^0 removal efficiency. It was realized by improving the transformation efficiency of Hg^0 into Hg^{2+} and Hg_p and then removing them by air pollution control devices. First, relationships between Hg^0 concentrations at the stack and variables like open values of secondary air, open values of over fire air, oxygen at the exit of economizer, load, coal qualities and so on were modeled with aid of tuned PCA-support vector machine. Then, manipulated variables and regulated variables were optimized by particle swarm optimization algorithm to enhance transformation efficiency of Hg^0 . A field thermal adjustment test was carried out on some 600 MW unit and the proposed method was applied to that unit and compared with ACO. Results showed that removal efficiencies were enhanced greatly in general. The increment of removal efficiency can reach up to 14.71%. Besides, optimal strategies can be found in few iterations, making it suitable for online applications.

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1. Introduction

Mercury is a trace element in coal. Coal combustion in power plants would result in mercury emissions characterized by persistence, long-distance transboundary transportation, bio-accumulation in the environment, and neurological health effects [1]. Coal combustion for electricity and heat generation utilization is the main source for anthropogenic mercury emissions [2,3]. In 2013, U.S. Environmental Protection Agency reinforced mercury emission limits for power plants, requiring that maximum emission concentrations should be in the range of 1.4 and 4.1 $\mu\text{g}/\text{m}^3$ (STP, dry) at 6 vol% O_2 [4]. Generally speaking, mercury emitted from coal combustion exists in three forms, namely elemental mercury (Hg^0), oxidized mercury (Hg^{2+}) and particulate bound mercury (Hg_p) [5]. Hg^{2+} is water-soluble, so it can be removed by flue gas desulfurization (FGD) scrubbers which are used for desulfurization [3]. Hg_p can be efficiently collected along with fly ashes by existing particulate control devices such as electrostatic precipitators (ESP) and

fabric filters (FF). However, Hg^0 is hard to be removed by existing devices [6].

Open values of over fire air (OFA), open values of secondary air (SA), oxygen at the exit of economizer (OE), and so on have great influences on the states of boiler [7]. Bai W et al. and Wang J et al. [8,9] found that tuning open values of SA and OFA can create high temperature and strong reducing atmosphere conditions in fuel-rich zone, which can help to reduce NO_x emissions and improve boiler's efficiency. Chen Z et al. [10] measured gas components, temperature, and so on under different stoichiometric ratios and recommended optimum ratios for economic, environmental, and safety operations. Hg^0 transformation efficiency at furnace back-end is just determined by the states of boiler. According to equilibrium calculations, mercury is only in the form of Hg^0 in high temperature [11]. Hg^0 is partially converted to Hg^{2+} and Hg_p via a series of homogeneous and heterogeneous reactions [12]. Smith CA et al. [13] found that HCl, SO_2 , and iron oxide particle surfaces were important contributors to mercury oxidation in flue gas. Yang Y et al. [14] found that mercury oxidation was sensitive to HCl concentrations. SO_2 weakly promoted heterogeneous mercury oxidation on fly ash surfaces, while the presence of H_2O showed an opposite effect. Xu W et al. [15] pointed out that big specific surface

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Nomenclature			
ACO	ant colony optimization	HOFA	high over fire air
CCOFA	close-coupled over fire air	OE	oxygen at exit of economizer
CEMS	continuous emissions monitoring system	OFA	over fire air
CVs	controlled variables	PCA	Principal component analysis
DVs	disturbed variables	PSO	particle swarm optimization
ESP	electrostatic precipitators	RVs	regulated variables
FF	fabric filters	SA	secondary air
FGD	flue gas desulfurization	SVs	state variables
GA	genetic algorithm	SVM	support vector machine
Hg ⁰	elemental mercury	T _{gas} ¹	flue gas temperature at exit of furnace
Hg ²⁺	oxidized mercury	T _{gas} ²	flue gas temperature at the stack
Hg _p	particulate bound mercury	MLP	multilayer perceptron
HS	harmony search	MSE	mean square error
		MVs	manipulated variables
		WFGD	wet flue gas desulfurization

areas and small pore diameters of fly ashes were beneficial for the mercury removal. Effects of fly ash compositions on adsorption and oxidation of Hg⁰ in flue gas were studied in details in Refs. [16,17].

In summary, open values of OFA, open values of SA, and OE determine the Hg⁰ transformation efficiency when coal qualities and the boiler's structure are given, because: 1) The states of boiler are determined by open values of OFA, open values of SA, and OE; 2) Hg⁰ transformation efficiencies are determined by the states of boiler. Bilirgen H et al. [18] carried out a thermal adjustment test to investigate impacts of boiler's modifications on Hg emissions. Investigated variables included OE, open values of OFA, burner and OFA tilts, and so on. Results showed that boiler modifications were beneficial for natural Hg reductions. Optimal combinations of boiler control settings resulted in a 34.5% reduction in Hg^T at the stack. However, optimal strategies offered by thermal adjustment method are only the best one of several attempts, which are worse than suboptimal solutions and the optimal solution. What's more, the thermal adjustment test is time and money consuming.

Combustion optimization based on the computational intelligence is a popular method to improve boiler's efficiency and cut down pollutants "naturally" in power plants [7], as it is effective, simple, fast, and economic. It is aimed to find the suboptimal solutions and even optimal solution. This method is composed of predicting model construction and optimization of manipulated variables [19–22]. Generally speaking, predicting model construction is based on machine learning methods like support vector machine (SVM) or neural networks (NN) and operation data containing substantial information of studied boilers. Optimization algorithms like particle swarm optimization (PSO), genetic algorithm (GA), ant colony optimization (ACO), or harmony search (HS) are applied to optimize manipulated variables based on the predicting model and related constraints. Applications show that 19.3% NO_x reductions can be achieved by this method for some studied case [22].

In the aspect of Hg emissions reduction, Jensen RR et al. [23] proposed a multilayer perceptron (MLP) to model the nonlinear relationship existing between mercury speciation and coal characteristics. Abdel-Aal RE [24] predicted mercury speciation based on a group method of data pretreatment, model synthesis, and selection of related inputs. Zhao B et al. [25] predicted mercury speciation based on SVM with flue gas temperature and coal qualities as inputs. Results showed that its performance was better than NN. Recently, NN has been improved and applied to predict mercury speciation again [26,27]. However, these studies only focus on the construction of predicting model. To the best of our knowledge, no study on combustion optimization based on

computational intelligence algorithms has been reported to reduce elemental mercury emissions in power plants. Our study proposes a computational intelligence approach to search optimal settings more comprehensively and faster. Compared with the thermal adjustment test method, this computational intelligence approach can offer better solutions to reduce Hg emissions.

The main contributions of this paper are:

1. A new combustion optimization technology for reducing Hg emissions in power plant is proposed for the first time. This technology is based on a predicting model of Hg⁰ concentrations based on tuned PCA-SVM and popular swarm optimization algorithm PSO. A comparative study is conducted along with thermal adjustment test and ACO. It can offer better operation strategies, namely controlled variables, than the thermal adjustment test method and ACO. Besides, it is very fast and can be used in supervisory information systems of power plants.
2. Variables in pulverized coal boiler system can be categorized into controlled variables and disturbed variables. Controlled variables are optimized while disturbed variables are kept fixed for a case to be optimized in the proposed method.
3. To explain why this computational intelligence approach can reduce Hg⁰ concentrations, optimized operation strategies are analyzed according to related research progresses of combustion theory and boiler technology.

The rest of the present paper is organized as follows. Section 2 describes the proposed method. Field thermal adjustment test of some 600 MW unit is given in Section 3. Then the proposed method was applied to the 600 MW unit. Results are shown in Section 4 and discussed in Section 5. Finally, Section 6 concludes this paper.

2. Methods

2.1. Reduction of Hg⁰ concentrations at the stack based on PSO

Variables in the pulverized coal boiler system can be categorized into four kinds. Variables like open values of OFA and SA can be manipulated by operators, so they are categorized into manipulated variables (MVs). Variables like OE are regulated by adjusting other variables like fuel amounts, air amounts and SA amounts. They are categorized into regulated variables (RVs). MVs and RVs are both named as controlled variables (CVs) for the convenient expression in this paper. For a given condition, the load and coal qualities are categorized into disturbed variables (DVs) as they cannot be adjusted. Flue gas species concentrations (initial Hg⁰, HCl, H₂O, NO_x

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