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Full Length Article

Modeling of mercuric chloride removal in ductwork and fabric filter by raw activated carbon injection

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HIGHLIGHTS

- Model for $HgCl_2$ capture by raw AC sorbent in ductwork and FF was developed.
- Adsorption kinetics/equilibrium, mass transfer and operating parameters considered.
- Internal mass transfer, sorbent size and loading determine in-flight capture.
- Sorbent size is the most important parameter in FF capture.
- Pressure drop for 20-μm fly ash is manageable up to 90-min cleaning cycle.

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ABSTRACT

Based on previous experimental adsorption kinetic and equilibrium data for mercuric chloride ($HgCl_2$) adsorption onto raw activated carbon (AC), a comprehensive model was formulated for its removal in the ductwork and fabric filter by raw AC injection in coal combustion flue gas. Various factors that can impact the performance and sorbent utilization were investigated, including inlet $HgCl_2$ concentration, sorbent loading, particle size, external and internal mass-transfer considerations, residence time, filtration time, injection mode, and pressure drop. Despite these considerations, in-flight removal of $HgCl_2$ in the ductwork was found to be almost negligible. For the removal in fabric filter, the effect of particle size was found to be significant, particularly for continuous sorbent injection. On the other hand, a discontinuous injection mode delivering the same amount of sorbent in 10% of a cleaning cycle resulted in higher removal performance and sorbent utilization. However, at the end of the cleaning cycle, most of the sorbent capacity was not used (<0.2%). The pressure drop across the filter cake built by fly ash and sorbent was found to be manageable within typical operating limits.

1. Introduction

The recent Mercury and Air Toxic Standard (MATS) rule limits the emissions of mercury (Hg) from power plants across the United States [1]. Under the MATS rule, the U.S. Environmental Protection Agency (U.S. EPA) started to reduce the mercury emissions from coal-fired power plants in 2015. Existing electric utility generating units (EGU) would limit their emissions below 0.18–18.14 g/GW h while newer EGUs would have stricter standards of limiting their emissions below 0.04–18.14 g/GW h depending on the type of fuel the EGU utilizes [2]. Activated carbon injection has shown promise in controlling gaseous mercury emissions in the particulate matter control devices (PMCDs) [3,4]. Previous model results suggest that the removal of oxidized mercury (Hg(2+)) in an entrained flow, using powdered activated carbon (PAC) sorbent is insignificant due to the mass-transfer resistance associated with the sorbent and its short residence time [5,6]. It was also demonstrated through the U.S. Department of Energy's (U.S. DOE) Field Test Program that the in-flight removal of mercury vapor with short residence times was insignificant [7]. In contrast, the removal of mercury in the PMCDs such as fabric filter (FF) and electrostatic precipitator (ESP) was reported to be significant [6–8].

It has been reported that raw activated carbon (AC) does not physically adsorb elemental mercury (Hg(0)) vapor in a typical post-combustion temperature window (e.g. 60–170 °C) [5–9]. Halogenated AC has shown good potential to increase the removal of both Hg(0) and Hg(2+) species [10,11]. However, little has been reported about the capture performance of each mercury species by AC sorbent in terms of the physical and chemical properties of AC such as surface area, porosity, reactions kinetics, and adsorption

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Nomenclature

C	$HgCl_2$ concentration inside the pore (g $HgCl_2/m^3$)
C_B	$HgCl_2$ concentration in the bulk gas phase (g $HgCl_2/m^3$)
C_B^{in}	inlet $HgCl_2$ concentration in the bulk gas phase (g $HgCl_2/m^3$)
C_B^{out}	outlet $HgCl_2$ concentration in the bulk gas phase (g $HgCl_2/m^3$)
C_i	inlet dust concentration (lb/ft^3)
D_e	effective pore diffusion coefficient (m^2/s)
D_p	sorbent particle diameter (m)
K	equilibrium constant, $K = k_1/k_2$ (m^3/g $HgCl_2$)
k_1	adsorption rate constant (m^3/(g s))
k_2	desorption rate constant (1/s)
K_2	specific cake coefficient (1/s)
k_g	gas-phase mass-transfer coefficient (m/s)
m_{AC}	sorbent injection loading in flue gas (g/m^3)
m_{Ash}	fly ash concentration in flue gas (g/m^3)
L	filter cake thickness (m)
L_F	final cake thickness formed at a final filtration time (t_F)
ΔP	pressure drop across the filter cake (Pa)
ΔP_c	pressure drop due to filter cake (Pa)
ΔP_f	pressure drop due to filter (Pa)
S_E	residual drag due to fabric (Pa/(m/s))
q	$HgCl_2$ uptake onto sorbent (g $HgCl_2$/g sorbent)
q_{avg}	average $HgCl_2$ uptake onto sorbent (g $HgCl_2$/g sorbent)
$q_{\max}$	maximum $HgCl_2$ adsorption capacity (g $HgCl_2$/g sorbent)
R_p	radius of sorbent particle (m)
r	radial distance inside sorbent (m)
r_A	intrinsic $HgCl_2$ adsorption rate (g $HgCl_2$ adsorbed/(m^3 sorbent s))

$r_{A,obs}$	observed $HgCl_2$ adsorption rate (g $HgCl_2$ adsorbed/(m^3 sorbent s))
t	time (s)
t_F	final filtration time (s)
u	superficial gas velocity (m/s)
V	gas velocity (m/s)
z	axial filter cake thickness from fabric filter surface (m)
$*$	equilibrium state

Greek letters

ε_b	bed porosity
ε_p	particle porosity
ρ_p	sorbent particle density (g/m^3)
φ	in-flight dimensionless sorbent uptake, see Eq. (8)
ξ	in-flight dimensionless radial distance, see Eq. (8)
γ	in-flight dimensionless pore concentration, see Eq. (8)
γ_B	in-flight dimensionless bulk concentration, see Eq. (8)
τ'	in-flight dimensionless time, see Eq. (8)
φ	filter cake dimensionless sorbent uptake, see Eq. (14)
ξ	filter cake dimensionless radial distance, see Eq. (14)
λ	filter cake dimensionless axial distance, see Eq. (14)
γ	filter cake dimensionless pore concentration, see Eq. (14)
γ_B	filter cake dimensionless bulk concentration, see Eq. (14)
γ_B^{out}	filter cake dimensionless outlet bulk concentration
τ	filter cake dimensionless time, see Eq. (14)

kinetics and equilibrium. Since a typical mercury concentration in coal combustion flue gas is on the order of ppb, a minimum AC sorbent to mercury mass ratio must be much higher than its stoichiometric ratio (e.g., ~>10,000:1) to achieve >90% removal with a typical AC sorbent of a ~20-μm diameter [12,13]. However, it is also difficult to find previous systematic studies about the effects of sorbent particle size, surface area, pore diameter and adsorption equilibrium and kinetics that can greatly impact on mercury capture performances in the ductwork and PMCDs.

Depending on a level of coal combustion, some unburned carbon remains in fly ash often as a consequence of low-NO_x combustion systems. It has been reported that the mercury removal performances by fly ash carbons greatly vary with respect to various parameters such as surface area, carbon content, chlorine content in coal, coal types, surface functionalities on carbon [14–16]. One of the previous studies predicted that <5% of mercury could be adsorbed by unburned carbon fly ash [15]. Despite the wide performance variations, there is a consensus that the adsorption capacity of fly ash carbons are generally limited. A previous study reported that the capacity for $HgCl_2$ is ~10 times smaller than that of raw AC [14–17].

In this study, the removal of mercuric chloride ($HgCl_2$) as a model oxidized mercury species in the ductwork and fabric filter using a raw AC sorbent (i.e. Norit America's DARCO-HG) has been systematically studied by taking into account the physical parameters of the raw AC sorbent, the physical adsorption equilibrium and kinetics for $HgCl_2$, inlet $HgCl_2$ concentrations, sorbent injection loadings, and external/internal mass transfer. The impacts of these considerations on capture performances in the ductwork and filter cake are demonstrated. A consideration of the pressure drop across the filter cake is also presented for the sorbent injection in a typical range of fly ash loadings. Previous studies have shown the effects of flue gas constituents on the adsorption capacity of $HgCl_2$ onto activated carbon [18,19]. SO_2 was reported to have a negative effect on the sorption as it reacts with basic sites on AC. HCl helped in increasing the sorption capacity while NO_2 helped in promoting the negative effect of SO_2 on the sorption capacity of $HgCl_2$ onto carbon. However, these effects have not been quantified for the adsorption of $HgCl_2$ and thus have not considered in this study.

2. Kinetic model for $HgCl_2$ adsorption onto raw AC

2.1. Model assumptions

Physical adsorption is considered a main mechanism for $HgCl_2$ adsorption onto raw AC for sorbent injection after air preheater at ~120–160 °C [19]. The adsorption kinetics was based on the Langmuir theory for this study as the experimental adsorption kinetic data best fit this theory [20,21]. In a post-combustion entrained flow after sorbent injection, the adsorption of $HgCl_2$ onto sorbents comprises of mainly three steps: (1) the external mass transfer of bulk phase $HgCl_2$ to the outer surface of the sorbent; (2) the internal mass transfer due to pore diffusion inside the sorbent particle; and (3) the surface adsorption of $HgCl_2$ onto the inner pore surface of the sorbent. In this study, the adsorption of $HgCl_2$ onto AC sorbent has been modeled using the following assumptions:

(1) The flue gas in the ductwork and filter cake follows plug flow;
(2) The axial diffusion is negligible;