



Lower order representations of evolving particle size distributions for rapid gas-particle mass transfer simulations during electrostatic precipitation

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ABSTRACT

The range of particle sizes typically present in combustion flue gas complicates predictions of gas-particle mass transfer processes. This complexity is amplified within electrostatic precipitators where particle motion, abundance, and mass transfer characteristics are size dependent. The present study illustrates the utility of replacing explicit representations of particle size distributions in simulations of electrostatic precipitators with an equivalent loading of monodisperse aerosols of diameter chosen to reproduce the same gas-particle mass transfer characteristics. Computational times are reduced by an order of magnitude using this approach, facilitating future incorporation of multiple particle types or heterogeneous chemical kinetics.

1. Introduction

Particulate control devices have historically been the most essential of emissions control processes employed at coal-fired power plants. Of these, electrostatic precipitators (ESPs) are the most common. Electrostatic precipitation research has been the subject of decades of research seeking to understand and predict collection efficiency [1,2], electrical discharge phenomena [3,4], the influence of gas and particulate matter properties [5–8], and electro-hydrodynamically induced flow phenomena [9–12]. More recently, as countries seek to reduce mercury emissions into the environment to adhere to the Minamata Convention on Mercury, coal-fired power plants become essential to achieving these goals. Since the mid-1990s, researchers have sought to understand the complex processes responsible for what has been found to be highly variable and site-dependent mercury emission rates during coal combustion. The widespread use of electrostatic precipitators (ESPs), combined with Hg control strategies relying on conversion of mercury from its elemental (Hg^0) and oxidized (Hg^{2+}) forms to particle-bound Hg (Hg_p), called for a better understanding of the intersection between the ESP phenomena and Hg adsorption onto suspended particles. Experimental data in this area is sparse, limited by the inaccessibility of the flue gas during treatment within an ESP and the difficulty in measuring trace concentrations of Hg in various forms. As a result, efforts to model the relevant underlying phenomena have largely been limited to the work of the present author and co-workers [13–17].

Mass transfer fundamentals predict, and more than a decade of research has shown, that increasing the interfacial area between the flue gas and suspended particles increases the adsorption onto particles of

mercury and other trace toxic metals present in the flue gas. In the case of elemental mercury Hg^0 , removal is a heterogeneous process requiring both Hg^0 mass transfer to a particle surface followed by oxidation from Hg^0 (to, e.g., Hg^{2+}) prior to or in conjunction with adsorption onto the particle surface. Similar principles govern mercury removal within gas-liquid contactors or spray dryer absorbers, installed to remove acid gases but also used to remove mercury from flue gas.

Accurately representing the interfacial area between a continuous phase such as the flue gas and a dispersed phase such as particles or droplets is essential to accurately predicting the mass transfer between them. In the present context of emissions control devices used in coal combustion, the characteristics of the interfacial area between continuous and dispersed phases falls along a continuum of complexity having two extremes. Gas-Liquid contactors used to remove acid gases can (though not always) fall nearer the lower end of this continuum, designed in multiple stages in which droplets are injected to meet desired size specifications, droplets large enough to fall downward due to gravity through an opposing upward gas flow. Within a single stage, moisture-saturated conditions prevent droplet evaporation, fixing droplet size. The opposed nature of the gas flow and droplet trajectories means that virtually all droplets are collected after traveling the same distance from their point of introduction to the flow. Under such constraints, the interfacial area between the droplets and the flue gas can be reasonably approximated as constant within each stage.

At the other extreme are ESPs tasked with removing mercury or other trace pollutants from flue gas. In this case, the dispersed phase consists of native fly ash containing varying amounts of unburned carbon and may be supplemented with powdered sorbents added to

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adsorb mercury. The fly ash and the sorbent have distinct particle size distributions initially and the total interfacial area available for mercury adsorption is the sum of their two contributions. However, the particle collection process is such that particles are removed continuously in any given stage with the larger particles removed more rapidly than smaller ones. Because particle surface area scales as diameter squared, the regression rate of interfacial area for the dispersed phase is different between different electrical sections, and is non-linear with time/distance within each section. The complexity and computational expense of simulating both particle precipitation and trace pollutant adsorption with ESPs is a barrier to the validation and optimization of approaches for reducing mercury emissions from coal combustion. In the present study, computationally expensive particle size-resolved numerical simulations within ESPs are replaced by an approach in which an initial size-resolved solution is used to extract a lower order, mean representation of the evolving particle size distribution that, as a monodisperse particle suspension in an ESP, reproduces rates of gas-particle mass transfer from the fully size-resolved distribution. Numerical simulation results reveal the combination of hydrodynamic, aerosol, and electrostatic conditions within which the lower order approach accurately represents the fully size-resolved approach.

2. Materials and methods

The computational approach has been described in detail previously [13,18] and therefore is only briefly summarized here. A synopsis of the approach is as follows: For a defined ESP channel segment geometry, solve for the electric field and space charge density distributions; assign entering particles a charge based on their size and the determined average electrical conditions within the channel segment; solve for the fluid and particulate motion throughout the channel based on 2-D turbulent Navier-Stokes equations including electric body forces; calculate cumulative rates of gas-particle adsorption based on mass transfer correlations for submerged spherical particles and local adsorbate concentrations. Two-dimensional representations of three- and nine-wire sections of a wire-plate ESP channel measuring 2 m × 0.3 m (L × W) and 5 m × 0.3 m, respectively, form the computational domain. The wires (1 mm diameter, 0.5 m spacing) represent discharge electrodes located along the channel centerline between the channel walls representing the electrically grounded collection electrodes. Previous analyses [13] and simulations [18] indicate that mercury adsorption by sorbent-covered solid surfaces is a small contributor to the total mercury removed in an ESP, and is therefore neglected in the present study.

In the computational domain, assuming the electric potential is not affected by the disperse phase, an external iteration [18] is used to determine the parameters A and n of an assumed distribution of space charge density q_i around each wire discharge electrode of radius R of the form $q_i = A(R/r)^n$ that is required to satisfy Poisson's equation and current continuity subject to the applied boundary conditions in Table 1. The 2-D, incompressible turbulent fluid flow field is solved for using a modified form of the Reynolds-averaged Navier-Stokes equations to include an electric body force term (Table 1) the spatial distribution of which depends on the solution for the spatial distribution of the electric potential. The default turbulence parameters are used. The resulting velocity field provides the initial condition for the turbulent two-phase flow mixture modules, facilitating a converged solution.

The initial log-normal size distribution of the dispersed phase entering the computational domain is represented in COMSOL™ by 11 separate two-phase flow mixture modules corresponding to the 11 particle size bins, from 1.5 to 125 μm, into which the distribution has been divided (Table 1). The 11 mixture modules solve for the spatial distribution within the domain of the number density or volume fraction of each dispersed phase size category due to the drift or migration of the charged particles in response to the local electric field and their

ultimate loss as they are collected on the planar collection electrodes. Particle motion relative to the continuous phase (particle slip) depends on viscous and Coulombic forces. The expressions for the x- and y-components of \vec{U}_{slip} , u_{slip} and v_{slip} , are (Eqs. 1 and 2):

$$u_{slip} = \frac{N_e(-e)E_x C_c}{3\pi\mu d_p} \quad (1)$$

$$v_{slip} = \frac{N_e(-e)E_y C_c}{3\pi\mu d_p} \quad (2)$$

where N_e is the number of elementary charges on each particle, e is the elementary charge of an electron (1.6E-19C), E_x and E_y are the x- and y-components of \vec{E} called from the electrostatics module, and μ and d_p are as previously defined. C_c is the Cunningham slip correction factor (Eq. 3):

$$C_c = 1 + Kn \left[1.257 + 0.4 \left(\exp\left(\frac{-1.1}{Kn}\right) \right) \right] \quad (3)$$

where Kn , Knudsen number, is defined as λ/d_p and where λ is the gas mean free path evaluated as we have done previously [14]. An explicit calculation of particle charging would require a Lagrangian simulation of particles along their trajectories through the electric field of $\vec{E}(x,y)$. For simplicity, the present study determines particle saturation charge based on particle size d_p and the average magnitude of the electric field magnitude within the computational domain $|\vec{E}(x,y)|$. For particles larger than 1 μm, as are all particles in the present analysis, field charging can reasonably be assumed to be the dominant charging mechanism (Eq. 4) [19]:

$$N_e = \left[1 + 2 \frac{\varepsilon - 1}{\varepsilon + 2} \right] \frac{E d_p^2}{4e} \quad (4)$$

where $E = |\vec{E}(x,y)|$, ε is the particle relative permittivity (dielectric constant) [–] assumed to be equal to that of graphite for carbon-based mercury sorbents, and d_p and e are as defined previously. For a spherical particle, the Frössling equation (Eq. 5) relates the mean Sherwood number \overline{Sh}_d and associated mean convective mass transfer coefficient \overline{h}_m to its Reynolds number based on $|\vec{U}_{slip}|$:

$$\overline{Sh}_d \equiv \frac{\overline{h}_m d_p}{D_{ab}} = 2 + 0.552 \left(\frac{\rho |\vec{U}_{slip}| d_p}{\mu} \right)^{1/2} \left(\frac{\mu}{\rho D_{ab}} \right)^{1/3} \quad (5)$$

where D_{ab} is the binary mass diffusivity of a species in a dilute mixture, here assumed to be elemental mercury diffusing in air (3.4E-5 m²/s, taken from [14]) and μ , ρ , and d_p are as defined previously. Particle-particle interactions and two-way particle-fluid coupling are not considered. All 11 two-phase mixture modules are solved simultaneously to properly couple the local collective mass transfer rate for all particles to the local reduction in mercury concentration.

The chemical transport of dilute species module in COMSOL™ solves for the spatial distribution of a dilute species within the continuous phase using the computed spatial distribution of collective mass transfer previously obtained from the 11 two-phase mixture modules, subject to the assumed boundary conditions (Table 1). While sorbents are characterized by a finite mercury adsorption capacity, characteristic times over which particles remain suspended in the gas flow within an ESP are comparatively short. When paired with the high mercury sorption capacities of modern, chemically modified sorbents, it is reasonable to treat the particles as perfect Hg sinks on whose surface Hg concentration is consistently negligible.

Existing input files used to obtain previous solutions [18,20,21] in v4.4 of COMSOL™ were adopted for the present study, executed in v5.2a. COMSOL™ automatically meshes the computational domain based on physical scale, the boundary conditions applied, and the order of the PDEs being solved. Each input file is configured to solve for, in

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