

NUMERICAL MODELING OF DIRECT SORBENT INJECTION FOR SO_2 REMOVAL

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Abstract

A modeling study of steady, turbulent, reactive multi-phase flow in a duct injection flue gas desulfurization (FGD) system is reported in this paper. The stream-function/vorticity method was used in finite difference form. The κ - ϵ model was used for turbulence closure. A simple integrated formulation was employed to compute the behavior of sorbent droplets injected into the flue gas stream. The sorbent may be injected either as slurry or solution into a hot flue gas, or a dry sorbent may be injected into a cooled and humidified flue gas. A comprehensive heat and mass transfer model was developed to simulate the evaporation of the sorbent droplets and the absorption/reaction of SO_2 in the sorbent droplets. Dissolution kinetics of lime particles within a slurry droplet was also included in this model to determine the overall SO_2 removal rate. Numerical calculations and comparisons with available experimental data were made and are discussed.

1. Introduction

In the United States, the emission of sulfur oxides has attracted much attention. The dispersion of sulfur oxides arising from combustion of fossil fuels has a dramatic impact on the environment. Removal of sulfur oxides from flue gases is very important in air pollution control. A sorbent in-duct injection system is proposed as a method for flue gas desulfurization. Chemically the system is similar to a spray dryer or dry scrubber.

In the sorbent in-duct injection system, a chemically reactive sorbent, usually lime or hydrated lime or dolomite, is injected into a highly turbulent flue gas containing sulfur dioxide. The sorbent may be injected as slurry or solution into the hot (typically 275 to 350° F) flue gas, or a dry sorbent may be injected into a cooled and humidified flue gas (typically 160 to 180° F). Processes that occur in the duct include the evaporation of water from slurry or solution spray with resulting cooling and humidification of the gas; sorption of sulfur dioxide into the water of the slurry or solution droplet; dissolution of the slaked lime into the water; reaction of the sulfur dioxide with the lime or dolomite; and some as-yet less understood effects, such as, the reaction of sulfur dioxide with "dry" sorbent after evaporation of slurry or injection of dry sorbent following humidification of the gas.

The Energy Conversion Research and Development Programs of the University of Tennessee Space Institute (UTSI) is presently involved in research of direct sorbent injection for combined SO_2/NO_x removal under Department of Energy Pittsburgh Energy Technology Center contract¹. Experiments are being conducted in the Direct Sorbent Injection Facility (DSIF). The objective of testing in this facility is to confirm the feasibility of a direct sorbent injection FGD system to control emissions of sulfur dioxide and nitrogen oxide from a fossil fuel plant. An extensive literature search also has been conducted at UTSI to collect and collate associated spray chemical reaction models.³ As a prelude to the development of an in-duct sorbent injection FGD system for commercial application by 1992, this literature search was carried out by UTSI at the request of Brookhaven National Laboratory and Department of Energy, Pittsburgh Energy Technology Center. The overall objective of the study was to assist the above organizations in ascertaining the state-of-the-art of computational methods applicable to the in-duct sorbent injection FGD system. This effort included analysis of existing analytical models, identifying salient features and shortcomings, and determining the ability of these models to be extended to adequately design and scale-up sorbent injection systems.

From the above studies, it is found that there is an extremely large gap between the needs of the design engineer in practice and the understanding and proposed research tasks of the academic researcher who desires to make a contribution on the FGD modeling front. Literature which describes numerical models for the sorbent injection FGD process is limited. Numerical models for predicting chemical reaction in multi-phase turbulent flow occurring in in-duct sorbent injection systems rest on two foundations: mathematical models of physical processes (turbulence, evaporation, absorption, reaction, and multi-phase effects) and computer codes for solving the appropriate level of sophistication of the resulting nonlinear governing differential equations.

Attempts to calculate the detailed performance of the FGD processes have only been undertaken during the last few years. Prior to 1986, the best computations available were based on overall global calculations⁴⁻⁹. The details of the interaction processes between the flue gas and the reacting, evaporating particle/droplet were not quantitative. Modeling of turbulent, reactive multi-phase flow in a spray dryer FGD processes is still in a state of development. Recent reviews of this subject for the FGD systems are given in reference materials.^{1,3,5,8,10,11} It has been determined that no model currently exists that will completely and accurately model the complex flow field and chemical reactions that take place in the sorbent injection system. However, models are available for portions

of the overall process. The available models that have been reviewed at UTSI are summarized in reference 3.

One objective of the FGD research at UTSI is to develop a numerical modeling for predicting steady, turbulent, reactive multi-phase flow in a spray dryer FGD system. In the SO_2/NO_x removal process development, a modeling technique using an adaptation of the LEE-2 computer program² has been employed at UTSI to simulate operation of the experimental program in order to delineate data that has been collected and to provide a route which leads to the accomplishment of design objectives. These efforts have included analyses directed at achieving an adequate model of the gas/particle dynamics and heat/mass transfer with or without chemical reaction for evaluation of experiments.

2. FGD Modeling Efforts

This section summarizes the numerical models which characterize the complex gas/particle flow field features for the direct sorbent injection in the FGD system. This work was undertaken to aid in evaluation of experimental performance of the DSIF test train. Its objectives were to provide a means by which the flowfields and the SO_2 removal process that develop within the FGD system could be viewed and qualified. Background on the model development and case studies that were performed using this numerical code in its original form are contained in the published works of Lee^{2,12-14}. For this present study, the model was refined to make it more amenable to the FGD duct. These refinements included a recasting of the governing flow equations to incorporate the effects of multi-phase flow behavior. A detail description of the model used is much too involved for any indepth discussion herein. However, a general overview of the model highlighting some of its techniques and salient features is briefly presented in following paragraphs.

The numerical model provides a solution to the elliptical, fully turbulent form of flowfield conservation equations in either a two-dimensional Cartesian or an axisymmetric coordinate system. The gas dynamic model which utilizes a stream function/vorticity formulation to the Navier-Stokes equations for compressible, turbulent flows forms the basis for the modeling. This approach provides a powerful means of analyzing recirculating flows wherein explicit dependency of the flow upon pressure is eliminated. Recovery of both the pressure and the velocity distributions throughout the flow is accomplished once the stream function and vorticity are defined from solution. The model utilizes the standard high Reynolds number form of the κ - ϵ model for turbulence closure.¹⁵ The model employs the law-of-the-wall and its related techniques to address near wall phenomena. This hypothesis allows realistic predictions of both wall shear losses and convective heat/mass transfer rates.

To solve the flow field equations in a non-equal spacing grid system a chain-rule technique is used to transform the physical spray-dryer plane into a rectangular computational plane. The governing equations and boundary conditions are rewritten in terms of computational plane coordinates and solved using finite-difference approximations. In order to extend the finite-difference equations beyond second order accuracy, a "decay function"¹⁶ is introduced. This technique assures stability in numerical manipulation of the equations at interior grid points of computational field. Solution of the finite-difference equations together with prescribed boundary conditions is achieved by an iterative, point by point, successive under/over relaxation Gauss-Seidel scheme.

The equations needed to model the particle or droplet trajectories are the differential equations of motion¹⁷. The particle trajectories are computed by integrating these equations, gravitational force, gas viscosity and gas velocity are assumed constant over the time of integration. The spray model involves mass, momentum and energy transfer processes which through their mutual coupling define the local state of the flow. This coupling can be either unilateral or bilateral, in other words, it can be assumed that the effects on the gas flow of the spray embedded within it are negligible, or, the effects of the presence of particles on the gas phase can be accounted for. The unilaterally coupled spray trajectory model is a useful first approximation to the overall behavior of a spray within a complex aerodynamic flow field. This technique becomes more exact as the spray becomes more dilute. Therefore, unilateral coupling is assumed for the spray trajectory model for the initial investigation. In order to employ this assumption, particle trajectories are computed by the equations of motion given the gas phase flow field for a variety of assumed initial particle sizes, velocities, and spray angles.

A heat and mass transfer model, SPRAYMOD, of SO_2 removal in a spray-dryer FGD system developed by Damle⁵ was employed to simulate the evaporation of the sorbent droplets and the absorption/reaction of SO_2 in the sorbent droplets. The code of SPRAYMOD was written in BASIC. This code was translated to FORTRAN for incorporation into LEE-2. The model is based on the assumption that the spray dryer could be simulated using a plug or backmixed flow model. Under this assumption, the effect of relative velocity between the flue gas and droplet is ignored. If the relative velocity is zero, the evaporation is the same as in a still-air condition. However, the initial velocity of the droplet at the exit of the nozzle is much higher than the surrounding flow. The effects of relative velocity on the simultaneous heat transfer from gas phase to the droplet, and mass transfer from the droplet to the gas phase have been considered during the constant rate of drying period. The particle/droplet trajectories of various droplet sizes and sorbent particle diameters superimposed in the two-dimensional turbulent flow stream have also been taken into account.

3. Results and Discussions

The numerical results performed in modeling of the DSIF are presented and discussed. A uniform gas velocity profile (Figure 1) adjusted in magnitude to satisfy mass conservation was used at the entrance to the test duct. Shown in Figure 2 is a 16" ID, 18' long cylindrical duct. Flue gas at 305° F enters from the left with a velocity of 10 m/s. The water particles at 110° F with a uniform velocity of 36 m/s enters from the left through a single water spray nozzle producing a 35° cone. The trajectories of the evaporating and reacting particles were calculated once the gas flowfield was obtained from solution. Figure 2 shows the trajectories of particles 25 μm in diameter superimposed in the gas flow stream. The shape of the particle trajectory is directly influenced by the magnitude and relationship between the drag forces and the buoyancy forces. A comparison to experimental data¹ obtained from the DSIF is shown in Figures 3 and 4. Therein it can be seen that the two-dimensional model for predicting this turbulent, two-phase flow is in near agreement with the laboratory experiments.

Figures 5-11 show the predicted SO₂ removal efficiency for both sorbent solution (sodium carbonate) and slurry (hydrated lime) cases. The results were computed subject to variety of sorbent particle diameters, dry sorbent reaction rate coefficients, inlet droplet diameters, inlet droplet temperature, inlet gas temperature, approach to saturation, and Ca/S stoichiometric ratio. The specifications for the spray dryer chosen as a reference case was based on the input conditions of DSIF SL00103A¹⁷. These are as follows: inlet gas temperature of 722 R, inlet gas velocity of 30 ft/sec, inlet droplet temperature of 600 R, inlet droplet diameter of 40 micron, inlet droplet velocity of 250 ft/sec, inlet SO₂ of 1171 PPM, a molecular weight of sorbent (Ca(OH)₂) of 74 lb/mole, sorbent density of 137 lbm/ft³, sorbent particle diameter of 4 micron, dry sorbent reaction rate coefficient of 1.0×10⁷, mole fraction of water in inlet gas of 9.3, approach to saturation of 29° C, and Ca/S stoichiometric ratio of 1.86. The discussion of these figures are given as following:

Effect of Inlet Sorbent Particle Size. Figure 5 shows that SO₂ removal efficiency decreases with increasing inlet sorbent particle diameter for the sorbent slurry. Smaller sorbent particles provide a larger cumulative surface area for reaction and would, thus, enhance the SO₂ removal efficiency. On the other hand, since the sorbent solution are highly soluble, the SO₂ removal efficiency is expected to be higher than that of sorbent slurry case. The sorbent solution case is shown not to be a function of inlet sorbent particle size.

Effect of Inlet Droplet Size. Figure 6 shows that SO₂ removal efficiency slightly decreases for the sorbent solution and slightly increases for the sorbent slurry with increasing inlet droplet diameter.

Effect of Dry Sorbent Reaction Rate Coefficient. The reaction rate coefficient depends on the diffusivity of SO₂ in the solid material, the particle size, and the moisture content of the particle/droplet.⁹ The ability of the chemical reaction model to predict observed efficiency is highly dependent on the user's choice of a reaction rate coefficient. Figure 7 shows the correspondence between the reaction rate coefficient and SO₂ removal efficiency for the set of operating conditions. This figure indicates that the reaction rate coefficient does not affect the efficiency until the coefficient exceeds a value of 10⁶ cm²/gmole.s. In the present study, a value of 10⁷ was assumed until further information becomes available.

Effect of Inlet Droplet Temperature. Figure 8 shows that SO₂ removal efficiency decreases very slightly with increasing inlet droplet temperature for both solution and slurry cases.

Effect of Inlet Gas Temperature. Several conclusions have been reported regarding the effect of inlet gas temperature on SO₂ removal efficiency. The experimental results performed by Apple and Kelly¹⁸ indicate that SO₂ removal efficiency increases with increasing inlet gas temperature. However, Buell¹ concluded from his experimental tests that the inlet gas temperature has a negligible effect. The numerical results performed by Damle, et al.⁹ and Ma, et al.¹⁰ indicate that SO₂ removal efficiency increases slightly with increasing inlet gas temperature. Figure 9 shows that SO₂ removal efficiency increases for the sorbent solution and decreases for the sorbent slurry with increasing inlet gas temperature. This implies that the effect of inlet gas temperature on SO₂ removal efficiency depends upon the form of sorbent.

Effect of Approach to Saturation. Approach to saturation is defined as the difference between flue gas exit temperature and dew point. Once the gas exit temperature and dew point are defined from the solution based on the process stream material and energy balance, the approach to saturation temperature can be computed. This is an important process parameter. Numerical results indicate that SO₂ removal efficiency decreases with increasing approach saturation temperature^{6,9} as illustrated in Figure 10. This is due to the decrease in total droplet area resulting from the decreased volume of spray during the drying period and the decrease in core volume of equilibrium water held by the solid after the drying period ends.

Effect of Inlet Ca/S Stoichiometric Ratio. Stoichiometric ratio is defined as the moles of Ca(OH)₂ fed to the system per mole of SO₂. This parameter is thought to be the most important factor influencing the FGD system performance. The overall SO₂ removal efficiency increases successively with increasing inlet Ca/S ratio.^{4,6,9,19} Figure 11 demonstrates this for both sorbent solution and slurry cases. This is due mainly to the decrease in liquid-phase resistance to SO₂ mass transfer during the drying period.

Finally, a comparison to DSIF experimental SO₂ removal data for this calculation is given in Figure 12. The

model predicted the SO_2 removal efficiency with a tendency to under-predict at the upstream of the spray dryer. Since no good correlations are available to estimate the liquid-phase resistance in the droplet to the mass transfer⁵ the difference between predicted and measured efficiencies was anticipated.

4. Conclusions

The numerical results presented above predict several significant phenomena in the FGD system. Although it is the author's opinion that a full understanding of these phenomena is far from being achieved, it is hoped that these results will aid in advancing the future research efforts. Major highlights and conclusions of the present study are:

1. A general two-dimensional computational procedure was developed to model steady, turbulent, reactive, multi-phase flow in a spray dryer FGD system. A chemical modeling program, SPRAYMOD, was successfully incorporated into a general fluid modeling program, LEE-2.
2. The effects of relative velocity on the simultaneous heat transfer from gas phase to the droplet, and the mass transfer from the droplet to the gas phase were considered during the constant rate of drying period. The particle/droplet trajectories superimposed in the two-dimensional turbulent flow stream were also taken into account.
3. Comparison with one set of the DSIF data shows good agreement between model predicted and observed particle velocity distributions.
4. Parametric studies reviewed herein have brought to light some of the gross effects of inlet-gas specifications and operating parameters on the duct injection FGD performance. The numerical results show that inlet Ca/S stoichiometric ratio remains the single most important aspect of the duct injection on its performance. However, other variables which showed appreciable influence included the dry sorbent reaction rate coefficient and the form of sorbent. This study provided a considerable insight for duct injection FGD performance optimization and scale-up.
5. To correct the tendency to under-predict the SO_2 removal efficiency at the upstream of the duct injection would require modeling of the wet particle stage.
6. A three-dimensional numerical model for a duct injection FGD system will be pursued as a future effort to compute the gas flow field, the droplet or sorbent particle dynamics, and the evaporation and chemical reactions simultaneously. The droplet/particle size distribution, agglomeration of particles, and wall deposition should also be considered in the model.

Acknowledgement

The author would like to thank Mr. T. E. Dowdy and Dr. J. M. Henry for providing valuable suggestions on the chemical model. The author would also like to thank Professor J. T. Lineberry for his assistance with this work. This work was sponsored by the U. S. Department of Energy under Contract DOE/PETC-DE-AC22-85PC81008.

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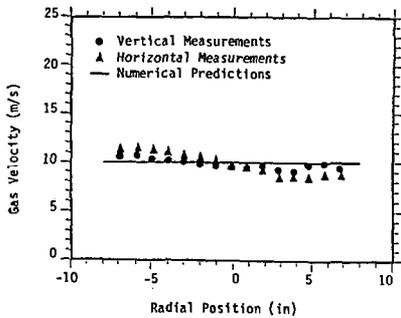


Figure 1. Gas Velocity Profile at Inlet to Test Section

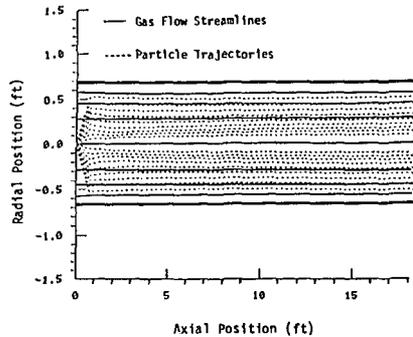


Figure 2. Particle Trajectories of $25\mu\text{m}$ in Diameter Superimposed in the Gas Flow Stream

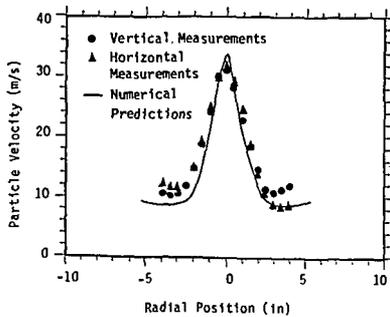


Figure 3. Particle Velocity Distributions, 18" Downstream of Spray Nozzle

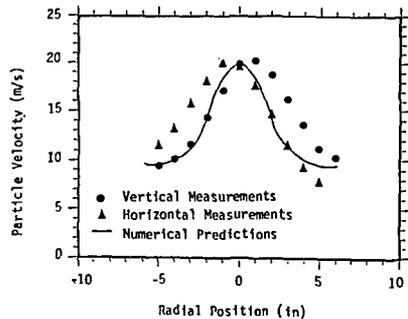


Figure 4. Particle Velocity Distributions, 36" Downstream of Spray Nozzle

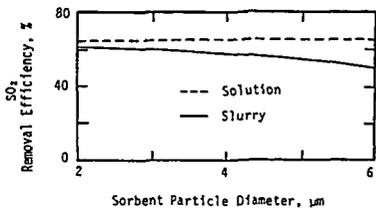


Figure 5. Effect of Inlet Sorbent Particle Size

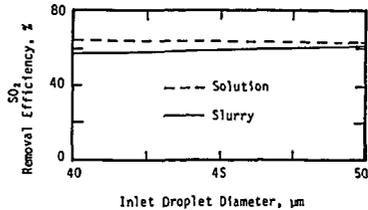


Figure 6. Effect of Inlet Droplet Size

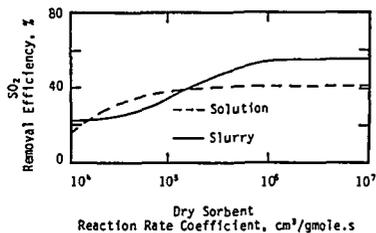


Figure 7. Effect of Dry Sorbent Reaction Rate Coefficient

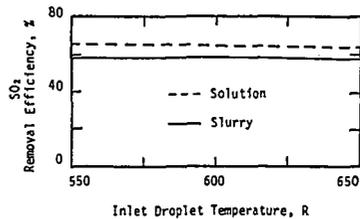


Figure 8. Effect of Inlet Droplet Temperature

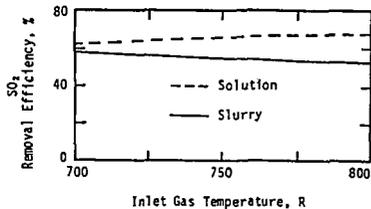


Figure 9. Effect of Inlet Gas Temperature

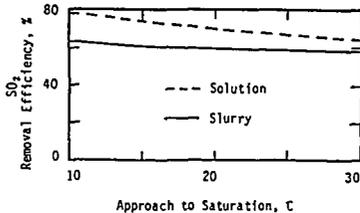


Figure 10. Effect of Approach to Saturation Temperature

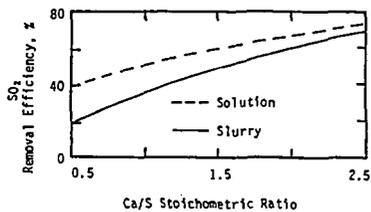


Figure 11. Effect of Inlet Ca/S Stoichiometric Ratio

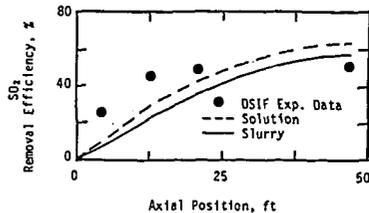


Figure 12. Calculated vs. Experimental SO_2 Removal Efficiency