PHYSICAL-CHEMICAL NO_x MECHANISMS IN
FLUIDIZED-BED COMBUSTION

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FLUIDIZED-BED COMBUSTION

Approved

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EXECUTIVE SUMMARY

The fluidized-bed combustion (FBC) of coal is being developed by the Department of Energy as an alternative coal conversion technology for increased utilization of the vast coal reserves in the United States. In addition to offering high efficiency at low temperature operation, the fluidized-bed combustor has the advantage of incorporating SO_2 reducing agents directly in the bed. It generally has been assumed that the operating low average bed temperatures insure low NO_x emissions. However, evidence indicates a potential for higher than expected NO_x emissions which threaten timely commercialization. This report details the effort undertaken at The Aerospace Corporation for the Office of Energy Research (Contract DE-AC01-79ER10027) to address NO_x emissions from fluidized-bed combustors and includes a summary of the important results obtained.

The approach adopted to develop a NO_x control strategy was to (a) identify the dominant physical-chemical phenomena which govern fluidized-bed combustor behavior and impact NO_x emissions, (b) employ physical models where needed which adequately describe those features, and (c) use the bounding estimates from the models to identify new NO_x mechanisms. Following this procedure, it has been possible to provide bounding estimates of combustion-related effects such as finite rate devolatilization and solids circulation, as well as outline the issues to be pursued in the development of NO_x control technology. The underlying premise which guided the selection process in (a) and the technology efforts under (b) and (c) above is that the evolution of NO_x in the combustion process is dependent on local conditions and thereby strongly coupled to the complex FBC fluid dynamics. Two aspects of FBC important in this regard are: (a) FBC operates in the large-particle (>1 mm) fluidization regime which is outside the range of most previous fluidization applications, and (2) up to 50 percent of the coal combustibles are comprised of volatiles which are released and burn in the gas phase. The unique characteristics of the large-particle fluidization regime, coupled with

the dense release of volatiles in the region where the coal is injected, contribute to a flamelike structure which plays a dominant role in FBC performance. Because this injector region structure determines local conditions for NO_x evolution, and because the volatiles themselves contain a significant fraction of the fuel nitrogen, volatiles evolution was selected as a focal point for this study.

A review of large-particle fluidization data as well as chemical time scales identified the need for a new combustor model to adequately describe volatiles evolution in the vicinity of FBC coal injectors. Such a model has been developed which treats the finite rate transport and devolatilization of injected coal particles as well as the gas phase transport and combustion of volatiles. This model differs from the previous generation 1-D models (averaged over the bed cross section) in its ability to predict spatial variations over the combustor cross section.

The model, as formulated, essentially describes the group combustion of dense gas-solid mixtures of devolatilizing coal injected into fluidized-bed combustors. Model calculations demonstrate the existence of three distinct combustion regimes in which: (a) the volatiles can burn in a large flame surrounding all the injected coal particles, (b) the volatiles can burn in a large flame surrounding a fraction of the injected coal particles while the remaining coal particles are surrounded by individual flames, or (c) the particles instantaneously release all volatiles upon entering the bed, and only gaseous volatiles are present which burn in a large flame. These predictions are a direct result of modeling the experimentally observed distinctive characteristics of large-particle (slow-bubble regime) fluidization which cannot be done with averaged one-dimensional bubbling bed models.

An important phenomenon associated with the group combustion characteristics of FBC is volatiles escape, whereby the dense volatiles flame does not consume all the volatiles within the bed. As a result, the volatiles escape into the freeboard above the bed where they burn, causing a shift in bed energetics. Experimental evidence of this phenomenon exists and is currently

regarded as a major design issue for FBC commercialization, aside from NO_x considerations. Volatiles escape was predicted with the injector region model, thus broadening the scope of the results to include implications for basic bed design.

Because the evolution of volatiles was the focal point of this effort, the important results can be viewed through their impact upon volatiles escape which strongly influences FBC design and scaleup. Results from the instantaneous devolatilization regime are used to benchmark the findings on the finite rate effects associated with the other two combustion regimes summarized as follows:

- Volatiles escape has been predicted to occur for a range of finite solids transport and devolatilization rates characteristic of commercial scale FBC.
- Volatiles release was found to increase over the instantaneous devolatilization limit by
 - More than 15 percent for finite solids transport and devolatilization rates
 - More than 25 percent for finite devolatilization rates and solids transport due only to convection with no dispersion
- Finite rate effects predict volatiles escape from combustors designed to eliminate it, based upon the instantaneous devolatilization limit.
- Finite heating times of injected coal particles enhance volatiles escape.

In general, it was found that finite rate processes enhance volatiles escape. The results on finite rate solids transport confirm that the large-scale solids circulation patterns, observed and studied for some time, do influence volatiles evolution and combustor performance. Therefore, the wide variations in observed solids circulation times must be resolved.

One of the most valuable uses of the present model, in addition to providing a design tool, is to guide the next step in experimentation and development based upon an accurate physical description of the important governing phenomena (i.e., large-particle, slow-bubble fluidization, dense devolatilization). Therefore, some recommended areas of investigation arising from the combustor model portion of the study are summarized here:

- Experimental and analytical investigation of freeboard dynamics including volatiles combustion and heat exchanger design
- Simultaneous determination of gas flow/solids circulation patterns and velocities
- Investigation of coal injector design and placement with respect to solids circulation patterns

Pursuant to these critical technology needs, a subcontract was let to Professor T. Fitzgerald of Oregon State University (OSU) to simultaneously determine the gas and solids flow patterns in the vicinity of a coal injector guided by the injector region model calculations. The staff of OSU were the first to identify the jetlike structure of gases injected into the bottom of a large-particle, tube-filled bed. This is a direct result of the unique character of the large-particle, slow-bubble fluidization regime and is the basis for the present model. The same apparatus has been modified to study solids injection. The first phase of this work has been completed and consists of data on gas flow and dispersion associated with a solids injector. It was found that the pneumatic transport gas which enters the bed with the solids does not flow in a uniform stream from the bubble cap type of injector. Instead, the gas flows out from under the bubble cap at a discrete circumferential location which varies randomly with time. This is an important result because it impacts (a) the initial particle heating rates which were shown to affect the level of volatiles escape and (b) the solids-gas contacting in the immediate vicinity of the injector and, hence, the initial group combustion rate. By incorporating these data in the combustion model, the sensitivity of volatiles evolution to this particular type of injector envisioned for commercial use can be quantified toward the next step in injector design.

With the first-order version of the combustion model completed, the implications of the injector region structure on NO_x evolution were investigated. New physical-chemical mechanisms associated with the predicted unique volatiles evolution structure were successfully identified, which are important to the understanding of NO_x generation and destruction in FBC. They are summarized here, with details given in the text of the report.

- Escaping volatiles carry a significant amount of fuel nitrogen into the freeboard, where conditions favor higher NO_x .
- Dense jetting volatiles in the injector region of the bed cause large excursions in air-fuel ratios.
- Jetting volatiles alter the volume of exposed char in the combustor available for NO_x reduction.

The first of these findings reiterates the need for freeboard modeling. It is important that NO_x considerations be part of freeboard design to achieve an optimum with respect to energy extraction and the completion of combustion. The last two mechanisms emphasize the importance of understanding the spatial structure as predicted by the present model.

Certain recommendations covering injector design, solids circulation data, and freeboard modeling were addressed in connection with the summary of finite rate effects on volatiles evolution. Additional recommendations cover extended modeling, those related to the OSU experiments, and those specifically directed at development of NO_x control technology. Based on the OSU experiments, it is recommended that the measurements of injected solids flow patterns be continued, and that the present model be used to determine the sensitivity of volatiles evolution to random injector gas flows observed. It is suggested, based upon the sensitivity of devolatilization to temperature for the residence times of interest, that a temperature calculation be added to the model. Temperature fluctuations associated with the gas voids should also be incorporated.

It is expected that only a NO_x model based upon the important physical-chemical phenomena of FBC will be adequate for design and scaleup. Therefore, the primary recommendation regarding NO_x emissions is that mechanisms

identified in this work be quantitatively assessed, based upon the injector region model. This is particularly important because the strong dependence of FBC NO_x characteristics on the complex fluid dynamics ties NO_x data interpretation directly to the fluid dynamic model employed. Much of the current wisdom in FBC NO_x emissions is tied to the one-dimensional averaged bubbling bed models. It is clear that this class of models cannot describe the volatiles flame structure, the volatiles escape phenomena, and, hence, the associated NO_x mechanisms. Therefore, it is appropriate that the whole issue of FBC NO_x characteristics be reviewed in light of the volatiles evolution structure discussed herein. A NO_x model based upon these recommendations will be tied to the governing bed phenomena and remain valid in the scaleup process. This is crucial in addressing expected reductions in allowable NO_x emissions from FBC which will require some form of multistaging. As such, the bed proper will be run with only a fraction of the required combustion air (substoichiometric), while secondary air will be injected into the freeboard to complete combustion. This will emphasize the role of the freeboard as secondary combustor, already made apparent by the volatiles escape phenomena. Therefore, the present approach to modeling the fluidized-bed combustor also provides an important link to the eventual design of multistaged FBC, where the emphasis will be on an accurate prediction of NO_x emissions.

1. INTRODUCTION

The fluidized-bed combustor (FBC), in addition to being an efficient low-temperature device, offers the major advantage of incorporating SO_2 reducing agents directly in the bed material for in situ control. Major design efforts have been expended in areas such as increased efficiency (e.g., fines recycle) and SO_2 control. Limited effort has been directed at NO_x control, based on the assumption that low average bed temperatures imply low NO_x . However, potential exists for higher than expected NO_x emissions. This could lead to the development of highly efficient FBC's which do not meet NO_x emissions requirements. To avoid this situation, it is necessary to develop an understanding of NO_x emissions as an integral part of FBC development. Because NO_x evolution is so strongly coupled with the combustion and fluid dynamic processes, it is appropriate for NO_x control to be developed in such a way.

The question of NO_x emissions in atmospheric FBC generally is neglected due to the belief that they are inherently low, based on the assumption that measured low average bed temperatures imply low NO_x emissions. This assumption must be viewed with care because:

- a. There is ample evidence that maximum local temperatures within the bed, which characterize thermal NO_x formation, are significantly higher than the measured average.*
- b. Eighty percent or more of the NO_x generated in coal combustion is through the conversion of fuel-bound nitrogen and is not a strong function of temperature, further suggesting low average bed temperatures to be poor NO_x indicators.**

*See Ref. 5

**See Ref. 23.

- c. There is evidence that the presence of sulfur may reduce NO_x emissions from coal combustion. As a result, in situ sulfur removal in FBC's may significantly increase NO_x emissions even in pressurized units.*
- d. NO_x levels have been measured** within FBC's showing large spatial variations independent of low average bed temperature.

In fact, the reason for the spatial variations noted in (d) is that high NO_x levels are generated in air-rich regions of the combustor followed by reduction due to the char- NO_x reaction.** This reaction is thought to be a dominant NO_x reduction mechanism in FBC whose strength increases with temperature. Therefore, the assumption that low average bed temperature engenders low NO_x emissions has little foundation in fact. Rather, it can be concluded that FBC NO_x emissions are a product of highly coupled, competing physical-chemical processes. They must be understood as a function of bed operating parameters in order to assure successful scaleup of commercial units while maintaining acceptable NO_x emissions.

It has been known for some time, based upon studies of oil and pulverized coal-fired utility boilers,[†] that conversion of fuel nitrogen is governed by local fuel-air ratio. As a result, the design of low NO_x burners for those applications is driven by controlling the air-fuel ratio seen by the fuel nitrogen species as they traverse the combustion zone. This emphasizes the need to understand the fluid dynamics of combustors. This same requirement exists for fluidized-bed combustors where local conditions throughout the bed must be known in order to specify the fuel-air ratio time history of coal particles and volatiles.

The approach taken in the present study toward a NO_x control strategy is to identify important bed phenomena which impact NO_x behavior. It then becomes apparent what modeling is necessary. Based upon modeling studies to

*J.O.L. Wendt and J.M. Ekmann, "Effects of Fuel Sulfur on Nitrogen Oxide Emissions," Proceedings of the Stationary Source Combustion Symposium, VI, EPA-600/2-76-152a, June 1976.

**See Ref. 23.

†O.W. Dykema, Analysis of Test Data for NO_x Control in Coal-Fired Utility Boilers, EPA-600/2-76-274, October 1976.

elucidate the particular NO_x -associated phenomena, it is possible to define areas, including experiments, requiring further attention. The key issue is that the modeling include an accurate description of the governing bed characteristics insuring that the NO_x emissions will be a predictable part of scaleup.

In view of the dependence of NO_x evolution on local combustion environment, attention was focused on bed fluid dynamics and associated combustion models. Fluidized bed combustors were found to operate in the large-particle ($> 1\text{mm}$) high-velocity ($> 1\text{ m/sec}$) fluidization regime which differs from most previous applications. This regime possesses certain unique characteristics which are not well represented by the general class of one-dimensional cross-sectionally averaged fluidization models. When combined with the fact that coal contains up to 50 percent volatiles, these characteristics foster a distinctive volatiles evolution structure in the vicinity of the coal injectors of bottom-fed atmospheric fluidized beds (AFBC). This injector region structure is comprised of dense volatiles released by the injected coal which burns in a large flame surrounding the injected coal. The injector region represents a large-scale dominant phenomenon which strongly influences FBC performance and plays an important role in scaleup. The group combustion characteristics of this region also determine the local conditions for NO_x evolution. Therefore, in conjunction with the approach adopted for the present study, the evolution of coal volatiles associated with bottom-fed AFBC was defined as the focal point. Therefore, the major effort of the work being reported was concerned with a group combustion model of dense gas/solids mixtures of devolatilizing coal being injected into the bottom of an AFBC. The details are given in Section 2. As a result of that work, critical development needs were defined in the area of solids circulation and injector design which impact both basic FBC design and NO_x emissions. A subcontract was let to Oregon State University (OSU) to pursue those issues with the results reported in a separate section on the experiments. The results of the model calculations also identified important new physical-chemical mechanisms influencing NO_x emissions which are also addressed in a later section of this report. Finally, conclusions are presented and recommendations made in the final section.

2. INJECTOR REGION MODEL AND FINITE RATE EFFECTS

The application of fluidization technology to the efficient and environmentally acceptable combustion of coal has brought to the forefront the large-particle (>1 mm), high superficial velocity (>1 m/sec) fluidization regime. The most notable feature of this fluidization application is that it operates in the slow bubble regime¹ such that the large-particle bed is characterized as turbulent-churning,² as distinct from the fast-bubbling, small-particle bed. The bottom-fed, shallow, AFBC envisioned for large utility scale applications is an example of a large-particle system. The special characteristics of such AFBC's contribute to a unique spatial structure which strongly influences performance and scaling. This spatial structure associated with the AFBC coal injector region and its dependence upon finite devolatilization and solids circulation rates is the subject of this report.

Figure 1 shows a typical commercial scale AFBC whose vertical as well as lateral dimensions are 1 m. Utility scale beds, although still restricted to the 1-m height, may be many meters in lateral dimensions with one coal feed/m². Therefore, Fig. 1 also represents the 1 m³ module upon which large-scale utility beds would be based. Upon entering the bed, the coal is transported by the solids circulation pattern, devolatilizes and, once devolatilized, undergoes char combustion. The volatiles account for as much as 50 percent of the coal heating value. Therefore, depending on comparative devolatilization and mixing times, regions of fuel- (volatiles) rich gas may exist in the vicinity of the injected coal giving rise to group combustion.

Whether or not group combustion occurs depends on how the volatiles are transported within the bed as they are formed. Some understanding of volatiles transport is provided by the experiments of Jovanovic³ which determined the gas flow patterns associated with the slow-bubble regime. Those experiments show that a tracer gas injected at the bottom of a tube-filled, large-particle bed rises as a plume. This is counter to observations in vigorously bubbling small-particle beds. These results suggest that volatiles released from injected coal rise through the bed as depicted in Fig. 1, burning in a diffusion flame with the surrounding oxygen.

Fig. 1. Bottom-Fed Atmospheric Fluidized-Bed Combustor

If the diffusion flame does not close within the bed, volatiles escaping into the freeboard will mix with air and burn, as first suggested by Park, et al.,² in an explanation of widely varying freeboard species measurements.⁴ Apparently, the spatial structure of the injector region resulting from the coupling of slow-bubble regime gas flow patterns and dense volatiles release strongly impacts the AFBC behavior and, specifically, the nature of the combustion therein. This underscores the importance of volatiles evolution and, in particular, the injector region as dominant phenomena in AFBC.

Early FBC studies⁵ focused on char combustion and neglected volatiles, greatly simplifying the modeling. Due to the importance of volatiles in the overall energy budget, recent studies have included some description of volatiles in models used. This was done by adopting various simplifying assumptions for devolatilization as well as solids/gas mixing rates. For example, Baron and Hodges⁶ assume the volatiles to be instantaneously released and uniformly distributed throughout the bed. Park, et al., in arguing that the general class of one-dimensional models based upon the two-phase theory of fluidization are inadequate for understanding volatiles evolution, assume that the volatiles are released instantaneously at the coal feed in order to focus upon the volatiles escape phenomenon. In most cases, these models are based upon arguments regarding the times scales of the pertinent physical-chemical processes. The present work is also based upon a review of the important time scales and concludes that finite devolatilization and solids circulation rates must be considered in the analysis of the injector region. The result is the ability to assess the importance of solids circulation for which Refs. 2 and 6 suggest significantly different time scales and, hence, vastly different modeling approaches.

The large-scale solids circulation patterns in large-particle beds have long been the subject of experimental research.⁷ However, the question remains as to the significance of solids circulation for the understanding of AFBC beyond noting that the solids are well mixed on the time scale of char combustion. The present work addresses this issue by quantitatively incorporating information on solids flow patterns through the solids circulation velocity.

The following subsection contains the formulation of the model including a review of the pertinent time scales discussed above. Subsection 2.2 discusses the results of the model calculations and their implications for AFBC design.

2.1 Injector Region Model

The complexity of the fluid-dynamic and chemical processes in the fluidized bed generally has dictated the need for simplified modeling with emphasis on those phenomena which most directly govern the bed behavior such as performance and scaling. The first such example was the formulation of the two-phase theory of fluidization⁸ which reflected the importance of the voids in determining such bed characteristics as mass transfer and gas/solids mixing. The importance of the injector region spatial structure in determining AFBC behavior discussed in the previous section suggests that it, too, is a governing phenomenon which must be considered in AFBC modeling. Models⁹⁻¹¹ developed around the two-phase theory of fluidization for small-particle applications generally cannot, without extensive modification, predict the structure of volatiles evolution in the injector region. There are two reasons for this. First, their transport mechanisms are representative of the fast-bubble regime. Second, they generally are one-dimensional (averaged over the bed cross section), whereas a multidimensional description is required to resolve the important features of volatiles evolution. As a result, such models cannot predict the volatiles escape phenomenon, the species variations in the freeboard, or the excursions of gaseous fuel-air ratios within the bed, which may have a bearing on the NO_x characteristics of AFBC.

To determine the best approach to modeling the injector region, the important physical and chemical time scales will be reviewed. They are listed in Table 1. Beer¹² discusses how these time scales are related to what is considered an optimum set of operating parameters for AFBC. It is evident that for AFBC temperatures (1100-1200°K) and particle sizes (1-10 mm), and hence overall particle residence times, the combustion times are large compared to the other time scales. This suggests the approach taken elsewhere¹² of assuming the devolatilized char particles to be well dispersed

and burning in the presence of oxygen. The gaseous volatiles are considered to burn instantaneously under diffusion-controlled conditions. The solids circulation time is a solids mixing time to be distinguished from the overall residence time of solids in the bed and is of the same order as the gas residence time. The comparison of solids circulation times quoted in Refs. 2 and 6 with devolatilization time scales¹² is the basis for the two completely different devolatilization models used in those investigations, as discussed earlier. However, it can only be concluded from Table 1 that the two processes occur on the same time scale and, hence, are coupled and must be considered in conjunction with the gas-phase transport and combustion processes. This is the motivation for including finite rate devolatilization and solids circulation in the model to be presented herein.

Table 1. Physical and Chemical Time Scales in FBC

<u>Physical</u>	<u>Chemical</u>
Solids Residence - >100 sec	Char Combustion - >100 sec
Solids Circulation - 2 sec (MIT) 10 sec (OSU)	Devolatilization - 1-10 sec
Gas Residence - <1 sec	Volatiles Combustion - <<1 sec (Diffusion-Controlled)

Before proceeding to the mathematical formulation of the model, a few words of clarification regarding solids circulation are in order. Solids circulation is comprised of both convective and dispersive transport. The manner in which the two combine to effect overall transport has a strong impact on chemical processes. That is, if a volume element is transported over a given distance with little mixing, then chemical processes are inhibited. Jovanovic's³ observations of injected tracer gas rising through a bed show, for example, that the wandering of the tracer plume resulted in an apparently augmented dispersion. However, it was noted that mixing was significantly

less on the scale important for reactions. Also, the quoted circulation times of 10 sec refer to the time for injected particles to become uniformly mixed throughout the bed. More important for reactions are the local velocities and dispersion rates in the vicinity where the injected particles are evolving spatially. The present model, by including finite solids velocities, mixing, and devolatilization rates, addresses this issue and is capable of assessing the comparative roles of transport and chemical mechanisms.

The physical quantities of interest in modeling the volatiles evolution associated with the coal injection process are the fraction of volatiles in the coal C_s , the gaseous concentration of volatiles released from the coal C_a , and the concentration of oxidizer C_b . Figure 1 sketches the evolution of these quantities in the AFBC. The volatiles containing solids are injected at the bottom of the bed, flow up through the bed at the solids circulation velocity, diffuse laterally, and devolatilize, thereby depleting C_s from its initial injected value. The gaseous volatiles C_a are produced by devolatilization, flow up through the bed, diffuse laterally, and are depleted by combustion with the surrounding oxygen C_b .

In order to model this process in a simple manner, it is necessary to make some assumptions to represent the important physical phenomena. The gases are assumed to be in plug flow with lateral diffusion, based upon the work of Jovanovic³ and Park, et al.,² in characterizing the slow bubble regime of large-particle fluidization. This is a key assumption which departs from the two-phase models based upon bubbles surrounded by clouds. The injected solids are assumed to be in plug flow at the solids circulation velocity in the vicinity of the injector. The solids also are assumed to diffuse laterally while undergoing devolatilization. Because solids mixing is more rapid than char combustion, the devolatilized coal particles are assumed to be well-mixed and burning in the presence of oxygen. The volatiles are assumed to undergo diffusion-controlled combustion at the interface between volatiles and surrounding oxygen. The particle and gas temperature are assumed uniform in order to simplify the initial calculations. In order to focus on the finite solids transport and devolatilization mechanisms, elutriation and char carryover are assumed to be zero.

Based on the foregoing assumptions, the injector region is modeled with a set of species equations for C_a , C_b , and C_s , expressing mass balances for the gaseous volatiles, the oxygen, and the volatiles remaining in the coal, respectively. The set treats the vertical flow and lateral dispersion of gas and solids while neglecting axial dispersion and is written as follows.

Volatiles in Coal:

$$u_s \frac{\partial C_s}{\partial x} = \frac{D_s}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C_s}{\partial r} \right) - W_d \quad (1)$$

Gaseous Volatiles:

$$u_0 \frac{\partial C_a}{\partial x} = \frac{D}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C_a}{\partial r} \right) + W_a^i + W_a^h \quad (2)$$

Oxygen:

$$u_0 \frac{\partial C_b}{\partial x} = \frac{D}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C_b}{\partial r} \right) + W_b^i + W_b^h \quad (3)$$

The symbols u_0 , u_s , D , and D_s represent the superficial velocities and dispersion coefficients of the gas and solids, respectively. The origin of the (x,r) coordinate system is at the centerline of the coal injector, with x measuring the distance above the distributor and r the distance

from the injector centerline. The injector radius at $x=0$ is denoted by r_0 and is determined by the coal mass flow and the solids circulation velocity u_s . The governing equations include reaction terms representing the devolatilization and combustion processes. The term W_d represents the reduction of volatiles in the coal C_s by devolatilization. The superscripts $()^i$ and $()^h$ refer to inhomogenous (solids) and homogenous (gaseous) reactions. Specifically, W_a^h is the loss of volatiles due to gas-phase combustion, while W_a^i is the production of volatiles that balances the depletion of C_s . Similarly, W_b^h and W_b^i are the oxygen losses due to combustion with volatiles and char, respectively.

Values for the gaseous dispersion coefficient are taken from Jovanovic,³ who observed that gas rising through the tube-filled bed dispersed due to particle scale rather than molecular motions and underwent a wandering motion probably due to bubble dynamics. The wandering motion, when averaged over long time scales, implies an apparent augmented dispersion; however, the particle scale dispersion controls the type of mixing important for reactions and is the appropriate choice for this formulation. Turbulent reacting flows exhibit a similar phenomenon in which turbulent motions apparently mix reactants on a large scale. However, it is actually the small-scale mixing which fosters reaction leading to the well-known thick turbulent diffusion flame.¹³ The same dispersion coefficient is used for both volatiles and oxygen, because the dispersion is controlled by the particle-induced gas motions rather than molecular diffusion.

Values of D_s are not as well-known as those for D . This partially explains the varying estimates of solids circulation times discussed earlier. In fact, one reason for the present work is to provide a tool for investigating the dependence of AFBC behavior on D_s . Although solids tracer studies in tube-filled, large-particle beds⁷ have not completely quantified D_s , they have suggested¹⁴ that dispersion is much smaller for solids than for gases. Therefore, D_s generally will be specified as some fraction of D and the effects of its variation observed.

The devolatilization process is highly complex and under study by many investigators.^{15,16} It is comprised of many orders of chemical bondbreaking requiring multistep kinetic mechanisms for accurate description.^{17,18} However, for the present purposes of exploring the role of such finite rate processes, it is adequate to use simple first-order global kinetics of Arrhenius type with reasonable numerical estimates as given, e.g., by Suuberg.¹⁶ This simplification is justified for AFBC temperatures because the complex, high heating rate, physical-chemical phenomena of coal-fired MHD¹⁵ are not in play. For the assumed uniform temperature fields, the kinetically controlled expression for W_d is

$$W_d = k_0 \exp\left(\frac{-E}{RT}\right) C_s \quad (4)$$

The source term W_a^i , representing a mass balance between the volatiles in the coal and the gaseous volatiles released by the coal, is related to W_d by

$$W_a^i = \frac{\rho_c}{\hat{M}_a} \left(\frac{\epsilon_{mf}}{1 - \epsilon_{mf}} \right) W_d \quad (5)$$

where ρ_c is the coal density, $\hat{M}_a$ is the molecular weight of the volatiles, and ϵ_{mf} is the voidage at minimum fluidizing conditions.

The consumption of oxygen by char combustion is given by

$$W_b^i = -k C_b \quad (6)$$

where $k = A_c k'$. The A_c is the char surface area per unit volume and k' is the effective char combustion rate coefficient related to the chemical rate constant k_s (m/sec) and the mass transfer coefficient k_g (m/sec) by the widely used expression

$$k' = \frac{1}{1/k_s + 1/k_g} \quad (7)$$

The value of k_s is taken from the work of Field¹⁹ as

$$k_s = 3.98 \times 10^4 T \exp \left(\frac{-35,700}{RT} \right) \quad (8)$$

while k_g is obtained from the Ranz and Marshall²⁰ Sherwood number evaluation.

Volatiles are considered to react with oxygen according to a simple irreversible one-step combustion mechanism

where a , b , d are stoichiometric coefficients and k_f is the rate coefficient. Of interest here is the fast reaction limit for which k_f is considered to be large. The gas phase reaction terms w_a^h and w_b^h are related according to the stoichiometry of the above reaction as

$$\frac{w_a^h}{a} = \frac{w_b^h}{b} \quad (10)$$

The boundary conditions for C_s , C_a , and C_b are

$$\begin{aligned}
 X = 0 \quad & C_a = 0 \\
 & C_b = C_{b0} \\
 & C_s = \begin{cases} C_{s0}, & 0 < r \leq r_0 \\ 0, & r_0 < r \leq L \end{cases} \\
 X > 0, r=0, L \quad & \frac{\partial C_a}{\partial r} = 0 \\
 & \frac{\partial C_b}{\partial r} = 0 \\
 & \frac{\partial C_s}{\partial r} = 0
 \end{aligned} \tag{11}$$

The analysis of equilibrium combustion problems of the present type is greatly simplified by the transformation²¹

$$\beta = b \left(\frac{C_a}{C_L} \right) - a \left(\frac{C_b}{C_L} \right) \tag{12}$$

where C_L is a reference concentration.

The governing equation set is transformed in the following manner. The equations for C_a and C_b are multiplied by b and a , respectively, and subtracted, after which the relation between w_a^h and w_b^h as well as the $(C_a, C_b) \rightarrow \beta$ transformation are applied. Finally, application of the coordinate transformations, $r^* = r/L$ and $x^* = x/H$, where L and H are the

lateral and vertical bed dimensions, respectively, results in the following equations for C_s and β :

$$\frac{\partial C_s}{\partial x^*} = \left(\frac{HD_s}{u_s L^2} \right) \frac{1}{r^*} \frac{\partial}{\partial r^*} \left(r^* \frac{\partial C_s}{\partial r^*} \right) - \left(\frac{H\Gamma}{u_s} \right) C_s \quad (13)$$

$$\begin{aligned} \frac{\partial \beta}{\partial x^*} = & \left(\frac{HD}{u_o L^2} \right) \frac{1}{r^*} \frac{\partial}{\partial r^*} \left(r^* \frac{\partial \beta}{\partial r^*} \right) \\ & + M \left(\frac{H\Gamma}{u_s} \right) C_s - S \left(\frac{Hk}{u_o} \right) \beta \end{aligned} \quad (14)$$

where

$$M = \left(\frac{b\rho_c}{C_L \hat{M}_a} \right) \left(\frac{\epsilon_{mf}}{1 - \epsilon_{mf}} \right) \left(\frac{u_s}{u_o} \right)$$

$$\Gamma = k_o \exp \left(\frac{-E}{RT} \right)$$

$$S = \begin{cases} 0, & \beta \geq 0 \\ 1, & \beta < 0 \end{cases}$$

The boundary conditions become

$$x^* = 0, \quad \beta = - (C_{bo}/C_L)$$

$$C_s = \begin{cases} C_{so} & 0 < r^* \leq (r_o/L) \\ 0 & (r_o/L) < r^* \leq 1 \end{cases}$$

$$x^* > 0, \quad r^* = 0, 1 \quad \frac{\partial C_s}{\partial r^*} = 0$$

$$\frac{\partial \beta}{\partial r^*} = 0 \quad (15)$$

The coefficient of oxygen consumption due to char combustion k in Eq. (14) is related to the char particle size through, for example, the char surface area per unit volume A_c . The char particle size, in turn, is part of the solution and generally is determined by a population balance as given by Levenspiel.²² Therefore, k must be specified as part of the solution. By assuming zero elutriation and carryover, the oxygen solution can be compared to a global oxygen balance on the bed, providing a means of iterating the solution to convergence by varying k . This simplification eliminates the need for a detailed consideration of char particle-size dynamics in the bed. It is important to account for the impact of char combustion on the oxygen distribution in the bed, because it determines the combustion rate of volatiles and, hence, the volatiles evolution. The global oxygen balance is expressed by

$$X_{O_2} = \left(\begin{array}{c} \text{Calculated excess} \\ \text{oxygen} \end{array} \right) = \left(\begin{array}{c} \text{Calculated oxygen} \\ \text{leaving bed} \end{array} \right) - \left(\begin{array}{c} \text{Calculated oxygen} \\ \text{to consume} \\ \text{escaping volatiles} \end{array} \right) \quad (16)$$

This value must be consistent with the excess air specified as a bed operating parameter.

Equations (13) and (14), and boundary conditions (15), govern the group combustion of a devolatilizing coal injected into an AFBC. The solution of those equations for a range of bed parameters defines the possible group combustion regime which can be observed in AFBC. These various regimes and their impact upon combustor behavior will be discussed below.

2.2 Discussion of Results

Three distinct combustion regimes were encountered in the solution of Eqs. (13) and (14). Which of these is actually observed depends upon the relative magnitudes of solids and gaseous transport as well as the devolatilization rate. Because the present model provides a framework for investigating the coupling of these competing phenomena, the remaining figures will quantify their role in determining AFBC behavior.

The basic calculations were performed for the bed geometry of Fig. 1 with parameter variation employed to investigate finite rate effects. A value of 0.04 was used for the parameter $HD/u_0 L^2$ based upon Jovanovic's data for D. The coal mass flow used is a 0.049 kgm/sec with a volatile matter of 43.7 percent. The value of excess air used in the calculation is 10 percent. The fraction of total combustion oxygen required for char combustion is 0.576. The bed temperature is 1116°K.

In addition to these parameters, the calculations shown in Fig. 2 employ a value for u_s/u_0 of 1/4 and for D_s/D of 0.01 in the range suggested by the work of Fitzgerald.⁷ For the devolatilization rate, $(H/u_s)\Gamma = 1.0$ is used. For this set of parameters, Fig. 2 shows typical radial profiles for C_s , C_a , and C_b at $X/H = 0.586$ selected at a convenient midheight computational station. The solutions are normalized by their centerline values for ease of plotting. The C_s profiles are of the expected form resulting from the devolatilization and diffusion process, as the solids are carried from the injection point by the circulation pattern. The C_a profile is the result of

Fig. 2. Profiles of Coal Volatiles (C_s), Gaseous Volatiles (C_a) and Oxidizer (C_b) at $X/H = 0.586$ for External Diffusion Flame

volatiles production and concurrent diffusion outward toward the diffusion flame where it goes to zero due to combustion. The C_b profile complements the C_a profile and falls to zero at the flame, confirming the requirement under fast combustion kinetics that fuel and oxidizer cannot coexist. This flame structure is termed "external," denoting that the gaseous diffusion flame resides outside the jet of volatiles containing solids.

Figure 3 displays calculations which differ from those of Fig. 2 by an increased diffusion ratio of $D_s/D = 0.125$. This variation is in the direction of increased circulation rates as suggested by estimates of Beer¹² in comparison with those of Park.² The important difference is that the gaseous diffusion flame has penetrated the solids jet. This is because the greater solids diffusion lowers the solids concentration in the outer regions of the C_s profile and, hence, the production of volatiles per unit volume. It is the volatiles production which balances the inward diffusion of oxygen. When volatiles production is large enough, it prevents penetration of the solids jet by oxygen resulting in the external flame of Fig. 2, while a reduced production rate permits oxygen to surround the devolatilizing particles in the outer edges of the jet as in Fig. 3.

A question exists as to the nature of combustion in the region outside the diffusion flame but within the C_s jet. The answer comes from close inspection of the solutions. The nonzero oxygen level in this region and the required noncoexistence of volatiles and oxygen are consistent with the absence of volatiles. Since the solids continue to devolatilize in this region, the only consistent interpretation is that the volatiles are immediately consumed by the excess available oxygen. This is an explanation within the context of the model of single particle volatiles combustion, in which the volatiles burn in a diffusion flame surrounding each particle. Beer²³ has observed such single particle volatiles combustion for coal particles dropped into a hot bed. The combustion zone for the "embedded" diffusion flame case therefore extends from the diffusion flame to the edge of the C_s profile, as indicated in Fig. 3.

Fig. 3. Profiles of Coal Volatiles (C_s), Gaseous Volatiles (C_a), and Oxidizer (C_b) at $X/H = 0.586$ for Embedded Diffusion Flame

The final combustion regime occurs for devolatilization rates which are large compared to solids transport, $(H/u_s)\Gamma \rightarrow \infty$, (D_s/D) finite. Figure 4 shows the calculations for this case in which the coal instantaneously devolatilizes upon entering the bed. Therefore, only gaseous volatiles are present in the bed. The combustion is once again confined to a diffusion flame, as shown by the solutions for C_a and C_b in Fig. 4.

Having established the group combustion regimes associated with dense gas-solid mixtures of devolatilizing coal found in the injection regions of AFBC's, it is of interest to consider the spatial structure shown in Figs. 5 through 7. Figure 5 shows a plot of the C_s jet and the diffusion flame surrounding the gaseous volatiles C_a in terms of (x^*, r^*) coordinates for the external flame case. Figure 6 shows the same plot for the embedded flame with the thick combustion zone indicated by a shaded region. These plots clearly show that the volatiles diffusion flames do not close within the bed, allowing the core of unburnt volatiles to escape the bed. The superposition of Figs. 5 and 6 in Fig. 7 shows that the relative position of the diffusion flame does not change very much. This raises the question as to the practical importance of distinguishing the two cases. The difference lies in the level of volatiles release, with the external flame emitting more volatiles. This is because the thick single-particle combustion zone consumes more volatiles.

Insight into finite rate effects upon AFBC behavior can be gained by investigating the interplay of the group combustion regimes. For example, Figs. 8 through 11 display finite rate effects on volatiles escape. Figure 8 shows the conversion X_v of gaseous volatiles for the instantaneous devolatilization limit as a function of $(HD/u_0 L^2)$, where X_v is the fraction of volatiles produced at $X = 0$ that are consumed in the bed. A value of $X_v = 1$ means that the volatiles flame closes within the bed and no volatiles escape. The parameter $(HD/u_0 L^2)$ is a ratio of convective residence to diffusion times. Since the gas combustion is diffusion-controlled, this parameter represents the ratio of residence to group combustion times and controls the degree of completion of combustion.

Fig. 4. Profiles of Gaseous Volatiles (C_a) and Oxidizer (C_b) at $X = 0.386$ for $\Gamma^a \rightarrow \infty$

Fig. 5. Flame Structure for External Gaseous Flame

Fig. 6. Flame Structure for Embedded Gaseous Flame

Fig. 7. Comparison of Embedded and External Flame Structure

If the assumption of instantaneous volatiles release were employed as a simplified design tool, then Fig. 8 could be used as a guide, for example, to eliminate volatiles escape. Furthermore, if increased bed height were employed as a solution, accepting the increased pressure loss penalty, a value of 0.12 for $(HD/u_0 L^2)$ might be selected as an optimum tradeoff. However, Fig. 9, which compares the finite and infinite rate solutions, shows that finite rate effects cause the volatiles escape to persist.

In the present analysis, uniform particle and gas temperatures have been assumed, implying that coal particles are instantly heated to bed temperatures upon injection. This assumption is reasonable, considering solids contacting heat transfer rates, but ultimately depends upon injector design and detailed solids flow patterns. However, it is possible to relax this assumption and provide a physical trend with respect to volatiles escape by specifying a temperature variation. The result shown in Fig. 10 is that volatiles production is not large enough in some cases to exclude the oxygen from the interior of the solids jet. Therefore, the complete cross section of the jet is in the single-particle combustion regime until the solids temperature reaches a threshold at which the increased volatiles production expels the oxygen from the interior region, thereby establishing a diffusion flame. Once again, the importance of this behavior for a given set of operating conditions is that finite heating rates can result in volatiles escape when not predicted by the instantaneous devolatilization limit. This trend is best understood by considering the conceptual limit in which it takes the full length of the bed for particles to heat up to a threshold at which the volatiles are immediately released and exit the bed unburned, resulting in maximum volatiles release. The calculation of Fig. 10 is an intermediate to this limit.

Finite rate effects are summarized in Fig. 11, wherein the concentration of escaping volatiles (at $x=H$) averaged over the bed is normalized with respect to the instantaneous devolatilization limit and plotted for finite devolatilization and solids circulation rates. The fraction of volatiles

remaining in the coal X_{cs} at $x=H$ also is shown. For $(H\Gamma/u_s) \sim 2.0$ and $D_s/D = 0.125$, which is in the range of parameters for the commercial scale AFBC of Fig. 1 operating at $T \sim 1100^\circ\text{K}$, the level of volatiles escape approaches 15 percent in excess of the infinite rate limit. For solids transport occurring only by the circulation velocity ($D_s/D \rightarrow 0$), the augmentation exceeds 25 percent. Note from the X_{cs} curve that the higher releases occur, even though the solids retain a portion of the volatiles.

Fig. 8. Conversion X_V of Gaseous Volatiles C_a at Bed Surface

Fig. 9. Finite Devolatilization Rate Effect on Closed Flame

Fig. 10. Flame Structure for Finite Heating Rate

Fig. 11. Unburnt Volatiles (C_a) and Coal Volatiles Conversion (X_{CS}) at Bed Surface

3. EXPERIMENTAL DETERMINATION OF INJECTOR-INDUCED FLOWFIELDS

Model results reported in previous sections clearly defined a need for experimental data on gas and solids flows associated with the solids injection process. This is apparent from the significant variations in volatiles release associated with the range of solids velocities and dispersion rates used in the calculations. Therefore, the model, by providing a formalism for including solids transport based on time scale arguments, has shown that the relative rates of convective and dispersive transport are important and must be quantified.

An experimental program was initiated at OSU on June 1, 1980, under subcontract to The Aerospace Corporation, in order to determine the required solids circulation velocities and dispersion rates as well as simultaneous gas flow patterns. The principal investigator was Professor T. Fitzgerald. He and coworkers first identified the jetlike structure of tracer gases injected at the bottom of a commercial scale fluidized bed. The same apparatus used for that work was modified to incorporate a pneumatic solids injector mounted through the distributor plate. It was intended to replicate injector designs envisioned for commercial use of the type presently employed in the EPRI-B&W pilot scale bed. As such, it can be operated at appropriate solid mass flow rates and scaled to the accepted design rule-of-thumb for number of feedpoints per unit cross-sectional bed area ($\sim 1 \text{ feedpt/m}^2$). Details of the apparatus and instrumentation are given in the Appendix which summarizes in report form the work done under the OSU subcontract.

The first step in the simultaneous characterization of gas-solids flows in the injector region is to investigate the gas flow. Since gas flows have been extensively characterized on this apparatus without a pneumatic solids injector, this work presented an excellent opportunity to observe injector effects on those gas flows. The tracer gas technique developed in the previous studies was also used here. This phase has been completed. It was

found that the solids carrier gas did not flow uniformly from the injector, which is essentially a vertical pipe with a disc placed normal to the pipe axis and some distance above the pipe exit. Rather, the gas flowed from under the injector cap at a discrete location on its circumference. This location moved around the circumference randomly. This type of injector gas flow has the potential for impacting both the initial heat transfer rates seen by the injected particles as well as the initial oxidizer diffusion rates to the volatiles flame. Model calculations discussed in previous sections show that these two factors have a marked influence on volatiles evolution and specifically on volatiles escape. The incorporation of this random process into the present combustor model would then provide a tool for assessing the impact of this element of injector design. For example, it would be possible to investigate the effects of altering the disc shape with the combustor model, again providing the link between cold model data and combustor performance. The data as well as the mathematical characterization of the injector gas flow as a random process are also included in the Appendix.

4. PHYSICAL-CHEMICAL NO_x MECHANISMS IN THE LARGE-PARTICLE, SLOW-BUBBLE FLUIDIZATION REGIME

With the unique characteristics of large-particle fluidization and dense devolatilization incorporated in a combustion model, it was possible to investigate the impact of those phenomena on NO_x emissions. As a result, new physical-chemical mechanisms were identified which are important to the understanding of NO_x generation and destruction in fluidized-bed combustors. These mechanisms are related to the structure of the volatiles evolution in the injector region. The volatiles are important in this regard, because they contain nitrogenous fragments originating in the fuel. The fuel-bound nitrogen, after some induction period, devolatilizes in proportion to the total volatiles. In fact, experimental evidence shows that after the volatiles other than nitrogen have been released, the nitrogen may continue to be driven off. Therefore, depending on the local residence versus devolatilization times, the coal volatiles may be enriched with respect to fuel-nitrogen. Additionally, the spatial structure of the volatiles evolution impacts NO_x by determining the local combustion environment (air-fuel ratio) in which the NO_x kinetics proceed, influencing not only the conversion of the nitrogenous species in the volatiles, but also any fuel nitrogen remaining in the char.

The first NO_x mechanism identified by the present model results is the transport of nitrogenous species into the freeboard by the volatiles escape phenomena. Because the volatiles can enter the freeboard before undergoing combustion, the nitrogenous species are likely to be unstabilized and in a form such as NH_3 . As a result, the conversion of these species will take place under vastly different conditions than those existing within the fluidized bed. For example, a major reduction mechanism in FBC, the char- NO_x reaction, which is associated with the bed of char in the combustor, will not impact freeboard nitrogen conversion. Only elutriated fuel would be available for reduction through this mechanism, while bed design is directed toward minimizing elutriation. The combustion of volatiles in the

freeboard in the absence of the dense particle heat sink of the bed results in temperature jumps above the bed. Therefore, the conversion of the nitrogen fragments in the freeboard is likely to occur under high temperature, locally oxygen-rich surroundings as compared to the interior of the combustor. These factors contribute to the possibility of enhanced NO_x emissions. More importantly, the fact that a certain fraction of fuel-nitrogen undergoes conversion in the freeboard may place a minimum on NO_x emissions beyond which further reduction may not be possible. Certainly, alteration of internal bed structure based on models which assume all NO_x mechanisms to proceed within the bed could not be expected to directly impact the above bed phenomena just described.

Additional physical-chemical mechanisms were found which impact NO_x evolution within the bed as a function of the dense volatiles flame structure. It is well-known that local air-fuel ratios along the path taken by nitrogen species in a combustor control conversion of fuel-nitrogen to NO_x . This is why it is important to understand fluid dynamic processes such as mixing. The volatiles flame structure described in some detail in previous sections results in large air-fuel ratio excursions across the lateral dimensions of the bed not predictable with one-dimensional averaged models. It is precisely these types of spatial inhomogeneities which drive the design of low NO_x , multistaged pulverized coal burners for the utilities industry. Any attempt at predicting the evolution of NO_x in the fluidized-bed combustor should incorporate these air-fuel ratio variations.

The volatile flame structure also affects the char- NO_x reduction mechanism in the bed. The inner part of the volatiles jet where only volatiles are present masks the available char from contact with any NO_x existing in the flame zone. This effectively reduces the available char bed for NO_x reduction. This must be taken into account when calculating NO_x destruction based upon carbon holdup in the bed so as not to underpredict NO_x levels. The reduction of exposed char bed also has implications for

backing char- NO_x kinetics out of fluidized-bed experiments. The attempt to do so is strongly dependent upon the fluid dynamics model employed. The use of a simple model which assumes that the char is uniformly dispersed and everywhere available for NO_x reduction will yield kinetics parameters which are a function of the model rather than the intrinsic kinetics.

Although more than 80 percent of coal NO_x emissions arise from fuel-nitrogen, the volatiles escape phenomena raise the possibility of some contribution from thermal fixation of combustion air nitrogen. This may arise from the high freeboard temperatures caused by the combustion of volatiles. Freeboard temperatures may be held down to 100-200°K in pilot scale units (~6 x 6 ft) with water walls and a sufficient number of feedpoints to minimize volatiles escape. In this case, freeboard heat exchangers can be placed well above the bed surface. However, for commercial scale units with much greater cross section, the ratio of effective water wall to cross-sectional area will be much lower requiring larger freeboard heat exchangers to be placed closer to the bed surface for both thermal protection and energy efficiency. If the heat exchangers are too far above the bed, the temperature jump can reach the adiabatic value of hundreds of degrees Kelvin, thereby boosting thermal NO_x levels. Rapid temperature drop above this zone due to heat exchangers can freeze the NO_x level with questionable effects on CO burnout. Alternatively, placement of heat exchangers directly in the freeboard combustion zone may hinder complete combustion. Consideration of NO_x mechanisms reemphasizes the need for careful engineering of the freeboard while placing additional constraints on that task.

5. CONCLUSIONS AND RECOMMENDATIONS

The results of an initial study to develop a NO_x control strategy for fluidized-bed combustion have been summarized in this report. The approach was to (a) identify the dominant physical-chemical phenomena which govern fluidized-bed combustor behavior and impact NO_x emissions, (b) develop a combustion model which adequately describes those features, and (c) use the model to define new NO_x mechanisms. Following this procedure, it has been possible to provide bounding estimates of combustion-related effects such as finite rate devolatilization and solids circulation, as well as outline the issues for executing the development of NO_x control technology. The underlying premise which guided the selection process in (a) and the technology efforts under (b) and (c) is that the evolution of NO_x in the combustion process is strongly coupled to local conditions through the complex FBC fluid dynamics. Therefore, it is necessary to base NO_x studies on a combustion model which accurately describes the unique characteristics of the large-particle fluidization regime in which FBC operates.

The body of the report has been organized into sections on the combustion model of the injector region, on experiments in support of model results, and on a discussion of NO_x mechanisms associated with the volatiles evolution structure predicted by the combustion model. The conclusions and recommendations will address those same general topics.

A combustion model was formulated to describe the spatial structure of the coal injection region including finite rate devolatilization and solids circulation. It was based on the observed unique fluidization characteristics of AFBC. Emphasis was placed on providing a simplified framework for assessing the impact of finite rate processes on volatiles evolution in AFBC by relaxing assumptions required in previous studies to include coal volatiles. This was done because the coal volatiles comprise a large fraction of coal-combustible matter and play an important role in both bed energetics and NO_x evolution.

The model as formulated describes the group combustion of dense gas/solid mixtures of devolatilizing coal. Three distinct group combustion regimes were delineated for FBC: (a) the external flame, (b) the embedded flame, and (c) the instantaneous devolatilization limit. The model correctly describes the single-particle mode of volatiles combustion observed by Beer,²³ which is responsible for the thick combustion zone structure of the "embedded flame." Calculations in all three regimes confirm the volatiles escape phenomena for commercial scale AFBC, thought to have been observed as freeboard temperature jumps.²

Model predictions show, in general, that finite rate effects have the potential for enhancing the volatiles escape phenomena. In particular, the model provides the mechanism for distinguishing between solids transport by dispersion and by circulation velocity, which can account for variations in quoted global circulation times.^{2,12} Calculations performed with varying ratios of solids dispersion and circulation velocity show a significant impact upon flame structure and volatiles escape. This suggests that it is indeed important to continue the study of solids circulation patterns. The present results underscore the importance of volatiles evolution and the spatial structure it fosters in the injector region as dominant phenomena in AFBC performance and scaleup.

Recommendations centering around the model development phase of this study are concerned with improving the model description of volatiles evolution and defining areas of experimental investigation. The sensitivity of devolatilization to temperature for the characteristic residence times of AFBC suggests the need to incorporate a coupled energy calculation. Temperature excursions associated with gas voids¹⁴ should be included and are amenable to treatment by certain statistical fluidization concepts.^{24,25} The parameter study for a range of solids dispersion to circulation velocity ratios showed a significant effect on the volatiles. This defines a need for quantifying solids transport in the injector region. An immediate consequence of the importance of solids flow patterns is the need to investigate their alteration and control as well as the placement of coal injectors. The impact of the simplified heating rate calculation on volatiles escape performed herein points to a need for the investigation of injector design.

The experiments performed under subcontract at OSU initiated a response to the recommendation that solids circulation and injector design be investigated. It can be concluded that the presence of the pneumatic injector of the type envisioned for commercial use does affect the gas flow in the injector region which has a potential for altering the volatiles generation and combustion. It is recommended that this work be pursued to completion by establishing the impact on solids circulation. It is further recommended that the impact of the gas flow results on combustor behavior be quantified by modifications to the model, which will define further needs for experimentation in that area. In addition, a number of other important experimental topics can be envisioned as a result of the present study, addressing issues such as optimum injector placement and design of the injector cap. The latter topic can be further refined with the recommended energy calculation by viewing the injector as a tool for tailoring initial heat transfer rates.

As pointed out in earlier sections, the unique character of volatiles evolution in AFBC has implications for NO_x emissions. The escaping volatiles can carry fuel nitrogen fragments into the freeboard, where their conversion is likely to proceed under oxygen-rich, high-temperature surroundings as compared with the interior of the combustor. Furthermore, the spatial structure of the injector region contributes to large excursions in air-fuel ratios known to influence fuel nitrogen conversion. Finally, the jetting volatiles alter the volume of exposed char available for NO_x reduction, which is suggested to be a dominant reduction mechanism in AFBC. With these mechanisms identified, it is recommended that their quantitative impact on NO_x emissions be assessed with the model presented here. The recommended model improvements can then be used to assess additional influence factors on the proposed NO_x mechanisms. For example, the treatment of temperature fluctuations will affect NO_x behavior indirectly by altering the volatiles structure while, at the same time, there may be a direct effect through the NO_x -char reaction which is believed to have a temperature dependence.

An important recommendation related to the volatiles escape phenomena is the development of a freeboard combustion model. The freeboard is known to be an important region; however, the present results emphasize the freeboard

as a secondary combustor wherein the completion of volatiles combustion takes place. As pointed out earlier, the conversion of fuel nitrogen in the freeboard must be given attention. Therefore, freeboard modeling is required both from the standpoint of bed energetics as well as NO_x emissions. A look toward expected future reductions in allowable NO_x emissions further emphasizes the urgency of understanding freeboard behavior. It is likely that attempts will be made to deliberately run FBC's in a multistage mode to meet the new standards. In doing so, the bed proper will operate substoichiometrically with secondary air injected into the freeboard. This can be construed as an extreme case of volatiles escape.

In summary, a significant step toward NO_x control technology for AFBC has been taken, based upon the framework of a relevant combustion model and the identification of important NO_x mechanisms to be investigated. In addition, results of this work directly benefit the basic process of AFBC design and scaleup. This in itself is important for NO_x control, because the structure of volatiles evolution in the injector region is a dominant phenomenon in bed scaling. Therefore, a NO_x emissions model should reflect the characteristics of that region if it is to be valid in scaleup. Another important result of this work is the methodology which has developed. That is, based upon the dependence of NO_x evolution on the fluid dynamics, a need for a combustion model was identified by considering experimental evidence of unique fluidization characteristics. A model was then constructed around the experimental data providing a formalism for assessing finite rate effects and the associated controversy. As a result, the finite rate effects were shown to be important, leading to experiments which can be fed back into the model which in turn can be used to define the next step in development. A key issue is that a combustion model of the present type provides a link between cold modeling (fluidized bed experiments in the absence of combustion) and combustor performance. This close feedback between model and experiment is the basic technique which should be applied to the general scaleup and development of fluidized bed combustion. The result is not only guidance in the development process, but ultimately a reliable design tool.

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Appendix
Experimental Investigation of Tracer Spread
in the Injector Region of Large Particle
Fluidized Beds

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INTRODUCTION

Most applications of fluidized beds involve gas fluidization of fine particles, usually less than 0.5 mm in diameter. Many investigators have researched gas flow, heat transfer and solids mixing in fine particle beds and have produced a number of very good relations. Several models based on the behavior of bubbles have been developed and used successfully to describe the performance of a fluidized bed as a chemical reactor.

Unfortunately, these correlations and models are not applicable for fluidized beds of large particles, and cannot be used to predict the behavior of such beds. Major differences in the fluid dynamic behavior of the beds of coarse and fine particles come from the different nature of gas flow through and around ascending bubbles.

Fluidization of large particles was not studied extensively until recently. Fluidized bed combustors typically use large limestone particles, greater than 1 mm in diameter, to scavenge the SO_2 produced by burning sulfur containing coal.

A properly designed atmospheric fluidized bed combustor should

- . Maximize the combustion of feed coal,
- . Minimize the production of pollutants, i.e., NO_x , CO , SO_2 , etc.,
- . Operate with a small percentage of excess air,
- . Minimize the utilization of limestone for sulfur capture.

These are competing requirements, and predicting the tradeoffs involved in different designs requires the use of some sort of model. Any general model which tries to incorporate all important factors will be complex because different physical and chemical processes occur in atmospheric fluidized bed combustors.

Physical and chemical processes that are important in modeling of atmospheric fluidized bed combustors are the flow of gases and solids in the bed, dispersion of feed solids away from the entry port, devolatilization of coal, combustion of volatiles in char, dispersion of flowing gases in the bed, etc.

Fluidized bed coal combustors will have unique features not found in other applications of fluidized beds. First, high gas velocities, essential in keeping the size of the bed within reason, require the use of large particles in the bed. At the same time the immersed tubes in the bed will restrict the growth of bubbles therein. Therefore the bed will be in either the slow bubble regime or in turbulent flow, but certainly not in the fast bubble regime.

Second, coal particles entering the bed will be in the high temperature region. Due to the high heating rate coal particles will release volatiles in a very short time. The behavior of the bed depends primarily on the time scales for devolatilization, for mixing and for combustion of coal. If the dispersion of feed coal were much faster than devolatilization, then each coal particle would have its own diffusion flame as its volatiles burn. However, the devolatilization is faster than the dispersion from a coal entry port and volatiles combine to form a large plume of unburned gas rising from the entry port. Since this plume contains roughly 50 percent of the heating value of the fuel it is not something which should be ignored.

Measurements show that the devolatilization time is less than 1 second for a 200 micron particle (Badzioch, 1961) and as much as 6 to 10 seconds for a 3,000 micron particle (Baron, et al., 1977); the particles require

15 seconds to be globally well mixed (OSU Progress Report, 1979); and the time required for the combustion of a charred particle is of the order of minutes (60 seconds for a 300 micron particle and 300 seconds for a 3,000 micron particle). Thus, one reasonable approximation is that most of the volatiles are released in the vicinity of the coal feed port. Figure 1 sketches the main features of the bottom fed coal burning atmospheric fluidized bed combustor. A typical concentration profile of volatiles and oxygen in the vicinity of the plume are shown in Figure 2.

Therefore, the major problem which remains to be solved for atmospheric combustion units is fuel introduction. It is a critical problem because combustion must be quite uniform throughout the bed. Gross fuel rich and fuel lean regions must be avoided. They would lead to lower fuel efficiency, difficult erosion problems and possibly combustion in the freeboard above the bed with a sufficient temperature rise to melt or sinter the ash produced by the coal.

Park, et al., 1980, address most of these problems in their efforts to set up a model for atmospheric fluidized bed coal combustors. The results of their modeling efforts could be synthesized in Figures 3 and 4. Both figures show that the conversion of volatiles and temperature jump above the bed are very strong functions of the dimensionless plume group $(HD/U_0)L_2^2$. This dimensionless group shows the effect of major design parameters, like bed height, superficial velocity, and coal feed port spacings, on the conversion of volatiles within the bed. However, all these parameters are not independent of each other, or at least cannot be arbitrarily chosen. The dispersion coefficient D is the most important parameter in the plume group since it is a function of characteristic fluidization velocity (see Jovanovic, 1979, p. 93) and

also limits the values of bed heights and spacings between feed points which could be chosen for a successful operation of the atmospheric fluidized bed coal combustor. Jovanovic, 1979 and Jovanovic, et al., 1980 elaborate on this subject. They show how one can experimentally obtain the values of dispersion coefficients in the large particle fluidized bed. They also developed the concept of meandering plume model which will be used for data analyses in this study.

The major goal of this study is to provide the data on dispersion coefficients in a scaled fluidized bed based on mean residence times of the volatiles in the bed. The effect of

- . different coal feed port configurations (one with the plate above the feed pipe and one without the plate)
- . different amounts of coal transport air, and
- . different superficial velocities

on the dispersion coefficient are also investigated.

CHAPTER 1

COLD FLUIDIZED BED MODELING; SIMULATION AND THEORY

Much work has been done recently using cold fluidized beds to investigate many of the characteristics of hot fluidized bed combustors. The advantages of cold fluidized beds are numerous; easier to build, easier to modify, easier to instrument and easier to change important design parameters. However, when hot fluidized beds are simulated by cold models great caution should be exercised. In order to have two different beds behaving similarly, in a fluid dynamic sense, several dimensionless groups should have identical values.

Four dimensionless groups are defined from the scaled fluidization equations given by Scharff, et al. 1977. These groups are

$$\frac{\rho_s}{\rho_f} \text{ the ratio of solid density to gas density} \quad (1)$$

$$\frac{d_p}{L} \text{ the ratio of particle size to a characteristic bed dimension} \quad (2)$$

$$\frac{u^2}{Lg} \text{ Froud number} \quad (3)$$

$$\frac{d_p u \rho_f}{\mu_f} \text{ Particle Reynolds number} \quad (4)$$

If these dimensionless groups are held constant when comparing two different beds, then both beds should have similar fluidization hydrodynamics.

If subscript 1 denotes the cold flow system and subscript 2 denotes

the hot flow system, then combining all the above dimensionless groups yields the following invariances:

$$L_1 = L_2 \left[\frac{\mu_{f1} \bar{\rho}_{f2}}{\rho_{f1} \mu_{f2}} \right]^{2/3} \quad (5)$$

$$d_{p1} = d_{p2} \left[\frac{\mu_{f1} \bar{\rho}_{f2}}{\rho_{f1} \mu_{f2}} \right]^{2/3} \quad (6)$$

$$U_1 = U_2 \left[\frac{\mu_{f1} \bar{\rho}_{f2}}{\rho_{f1} \mu_{f2}} \right]^{1/3} \quad (7)$$

$$\rho_{s1} = \rho_{s2} \frac{\bar{\rho}_{f1}}{\bar{\rho}_{f2}} \quad (8)$$

Combining L and u result in the scaling of time $t = \frac{L}{u}$ as

$$t_1 = t_2 \left[\frac{\mu_{f1} \bar{\rho}_{f2}}{\rho_{f1} \mu_{f2}} \right]^{1/3} \quad (9)$$

Scharff, et al. 1977, went into considerable detail describing an experimental plan for investigation of

- a) quality of fluidization,
- b) partial slanting, and
- c) dispersion of salts.

To test this theory Fitzgerald and Crane, 1980, conducted a set of experiments using two cold fluidized bed models with identical dimensionless numbers. The results of their investigation strongly indicate that the four dimensionless groups can be used for scaling of the cold models. That is,

the quality of fluidization of one cold model can be used reliably for prediction of the quality of fluidization of an exactly scaled second model even in extreme cases. They also concluded "it therefore seems likely that fluidization fluid dynamics of hot fluidized beds can be modeled in the scaled cold bed using the same dimensionless groups."

Following a similar line of reasoning and searching for the dimensionless groups which could best describe the quality of fluidization of gas fluidized beds, Jovanovic, 1979, developed a criterion for distinguishing between slow and fast bubble fluidization regimes. The criterion is based on the existing experimental and theoretical equations and is good for any particle size, density, fluid and temperature. Figure 5 summarizes the results from this effort.

CHAPTER 2

DESIGN OF COLD MODEL FLUIDIZED BED

The cold studies performed at Oregon State University are scaled to represent typical fluidized bed combustor designs used in current demonstration facilities.

Literature surveyed showed that a wide variety of bed material, particle sizes, tube arrangements and other experimental conditions are employed in different hot fluidized bed facilities. Some difficulties were encountered in choosing the material for the tube bundles and the bed media. These materials are simply not available in the sizes which would exactly correspond to the particular scaled sizes used in hot fluidized beds. For that reason the decision is made that the design of cold model fluidized beds should scale as close as possible, a typical design such as Babcock and Wilcox's 1.8 m (6.0 ft) fluidized bed combustor.

Still, design parameters of Babcock and Wilcox's hot fluidized bed could not and should not be scaled according to the dimensionless groups given in the previous section. For example, the size of the coal feed pipe used by Babcock and Wilcox should not be scaled according to geometric scaling factors, because the air in the coal feed pipe is not at the same temperature as the air in the bed. Therefore some compromises have to be made in order to preserve as close as possible the fluid dynamic similarities and quality of fluidization between hot and cold beds.

The hot flow system is gas (essentially nitrogen) at 871°C (1600°F) with a density of 0.32 kg/m^3 and viscosity of 0.047 cp . The bed consists of

limestone particles with a mean diameter of $d_p = 1208 \mu\text{m}$ filled to 1.2 m (4 ft) above the distributor plate and a tube bundle made of 2 in. tubes arranged in a triangular pitch as shown in Figure 6 (horizontal pitch to diameter ratio is 1.5, vertical pitch to diameter ratio is 1.67).

The cold flow system is air (also essentially nitrogen) at 37°C (100°F) with density of 1.15 kg/m^3 and viscosity of 0.0185 cp .

Table 1 summarizes all the important design characteristics of both, hot and cold fluidized beds and also shows calculated scaling factors. In order to find commercially available materials for tube bundles and fluidizing particles, a slightly different scaling factor had to be used.

TABLE 1. Some design characteristics of hot and cold fluidized beds.

Design Parameters	3&W	Scaling factors*	OSU
ρ_f	0.32 Kg/m ³		1.15 kg/m ³
μ_f	0.047 cp		0.0185 cp
bed height	1.22 m	0.229 (0.25)	0.3 m
d_t	0.05 m (2")	0.229 (0.25)	0.012 m (1/2")
$\bar{d}_p$	1208 μ m	0.229	302 μ m
ρ_s	2500 kg/m ³	3.59 (2.8)	7000 kg/m ³ (iron powder)
u_o	2.44 m/s (8 ft/s)	0.478	1.16 m/s
horizontal pitch to diameter ration	1.5		1.5
excess air in % of total gas flow	10% - 20%		10% - 20%

* numbers in parenthesis indicate assumed values for scaling factors.

CHAPTER 3

EQUIPMENT AND INSTRUMENTATION

Equipment

OSU's existing 0.91 m by 0.91 m square fluidized bed was modified to produce a one quarter scale model of Babcock and Wilcox's 1.83 m by 1.83 m hot fluidized bed located in Alliance, Ohio.

The 0.46 m by 0.46 m model contains a scaled tube array and a pneumatic center feed coal injection port. The fluidizing air, supplied by a roots blower powered by a 325 HP diesel engine, is distributed through a perforated plate. The coal transport/injection air is supplied by a separate smaller blower through a 2.54 cm diameter pipe located at the center of the distributor plate.

Figure 6 provides a comparison of OSU's and Babcock and Wilcox's test facilities and details of the sampling probe locations and Figure 7 shows the layout of the OSU test facility.

Instrumentation

Introduction

Gas tracer detection can be accomplished by a variety of means. The tracer gas must have flow behavior like the bulk fluid, but must have a measurable characteristic which distinguishes it. Physical properties such as heat capacity can be used for moderately large trace concentrations, but for low tracer gas concentrations radioactive or chemical tracers are generally used, because they can be measured accurately even at very low concentrations. Radioactive tracers have been avoided because of difficulties in handling them, particularly in disposing of them once they have gone through

the bed. Instead, a photo-ionization detector was used for monitoring tracer gas concentration. It behaves rather like a flame ionization detector: a carbon-containing compound is ionized--in this case by ultraviolet radiation--and the ion current is amplified by an electrometer amplifier and recorded. The response of the device is very rapid--on the order of a few milliseconds--and its sensitivity is high enough so that it is possible to use tracer concentrations of a small fraction of a percent by volume. Propylene is used as the tracer gas because it is inexpensive and gives a strong response from the photo-ionization detector. Under normal operating conditions (without careful cleaning), the photo-ionization detector is sensitive enough to detect a propylene concentration of .001% with a background reading that is less than 1% of the signal. The response time of the tracer injection system and the tracer sampling system is short enough (15 to 20 milliseconds) to allow transient tracer measurement in the bed.

Tracer Injection System and Sampling System

Propylene is injected into the bed along with the excess coal feed air through the center feed coal injection port. The excess air is regulated by a rotometer/venting valve system prior to the propylene mixing point. The propylene concentration is controlled by a pressure regulator and needle valve assembly and a reference probe located at the mouth of the coal feed port.

The tracer sampling system consists of: (a) three sampling probes; (b) one reference probe; (c) photo-ionization detector; (d) vacuum pump; (e) electrometer; and (f) low pass filter. A block diagram of the tracer sampling system is shown in Figure 8.

Gas is withdrawn through a sampling probe from the bed; it passes through

the photo-ionization detector and exits through a mechanical vacuum pump. The probe uses a porous stainless steel filter to prevent solids from being withdrawn along with the gas. Stainless steel tubing is used which has an outside diameter of 1.6 mm (1/16 in.) and an inside diameter of 0.076 mm (.003 in.). Rapid response of the sampling probe is obtained by maintaining a high gas velocity in the sampling line. This is accomplished by operating the photo-ionization detector at low pressure (0.066 atmospheres). In order to prevent leakage of air into the photo-ionization detector at this reduced pressure, the entire unit is enclosed in an air-tight vessel which is maintained at 0.066 atmospheres. The photo-ionization detector exhausts directly into this chamber. The pressure in this chamber is read on a closed-end mercury manometer.

Photo-ionization detectors behave much like a flame ionization detector, except that they are more sensitive and more stable. A wide variety of organic compounds and some inorganic compounds can be detected.

A schematic diagram of the photo-ionization detector is shown in Figure 9. The photons (10.2 eV energy) which are emitted from the lamp pass through an ultraviolet light transmitting window into the ionization chamber where absorption of the radiation by a molecule which has an ionization potential of 10.2 eV or less will lead to ion formation via the reaction:

A positively biased high voltage electrode is used to accelerate any ions formed by absorption of ultraviolet light to the collector electrode where the current (proportional to concentration) is measured.

If the carrier gas absorbs ultraviolet radiation strongly, another set of reactions may occur:

Either reaction can yield the same result if the energy transfer process is efficient. However, photo-ionization with 10.2 eV photons does not provide any sensitivity to permanent gases because of their high (> 12 eV) ionization potentials. Only species with ionization potentials below about 10.5 eV can be detected by the 10.2 eV source.

Photo-ionization is practically a non-destructive measuring technique. The apparent ionization efficiency is 10^{-4} , only 0.01% of the sample is actually ionized. The detector senses the concentration and as the sample flow rate is decreased, the sensitivity increases. The instrument has a nominal sensitivity of 1.5×10^{-2} microamperes when the concentration of propylene in the chamber is 10^{-4} atmospheres. The response is essentially instantaneous and is limited by the flow of gases through the tracer sampling system.

The ion current obtained from the photo-ionization detector is measured by an electrometer amplifier. The heart of the electrometer amplifier is a very low bias current operational amplifier (model 310) manufactured by Analog Devices. The amplifier is connected to measure the current in the photo-ionization detector as shown in Figure 10. The sensitivity of the amplifier is adjustable from approximately 5 to 100 volts per microampere input current. Thus a concentration of 500 ppm in the photo-ionization detector should produce an output voltage of (approximately) 7.5 volts at the high gain setting.

Careful shielding precautions are needed in order to reduce 60 Hz pickup.

All the resistors shown in Figure 11 are included in a small metal box on the back of the socket for amplifier.

The output signal from the photo-ionization detector for a steady tracer concentration is not smooth. It fluctuates 120 times per second due to the fluctuation in intensity of the ultra-violet lamp. The fluctuations in ion current which are caused by AC operated ultraviolet lamp are effectively removed by filtering the output circuit signal through an eight order Butterworth filter located on a small board in the same module as the electrometer amplifier. It has a DC gain of 1.33 which raises the output voltage to 10 volts when the propylene concentration in the photo-ionization detector chamber is 100 ppm atmospheric pressure. The electrometers used with the photo-ionization detector are housed in a small cabinet which contains its own power supply (Analog Devices Model 902). The output from each photo-ionization detector is connected through a separate BNC connector to its electrometer amplifier; the amplifier output is connected to the analog-to-digital converter at the minicomputer.

CHAPTER 5

MEANDERING PLUME MODEL

Predicting the performance of a gas-solid fluidized bed as a chemical reactor requires that we know the flow pattern of gas through the bed. Different approaches have been used to study the movement of gas. All of them, however, utilize some type of tracer gas as a tool for exploring the flow characteristics. Steady-state and unsteady-state tracer injection are the two possible techniques of introducing the tracer gas. The interpretation of the experimental results may take different routes.

Fitting data to some type of diffusion model is probably the simplest and most widely used approach. Axial and lateral dispersion coefficients are the parameters of these models. Several mathematical techniques are used to solve the basic diffusion equation whose complexity depends on the boundary conditions and simplifying assumptions.

Stimulus-response curves are often used to interpret flow patterns in a fluidized bed. Levenspiel, 1972, gives a detailed explanation of this experimental technique. F and C curves are useful for the evaluation of a mean residence time of gas and the fluidized bed as a whole. However, if we want to learn how long a gas element spends in the emulsion or in the bubble phase, we have to resort to more elaborate and complicated tracer techniques. The stimulus-response technique fails to give a unique answer in this type of application, unless they are coupled with other information about the investigated phenomena.

The results of tracer experiments are also used to fit bubble-emulsion models. A number of two-region models have been proposed but only a few of them have survived the test of practical application. Caution has to be exercised

when tracer results are used to determine the parameters of these models. Some two-region models are "loaded" with a large number of adjustable parameters. The excellent fit, usually obtained with these models does not prove the validity of the model. It is rather a consequence of the large degree of freedom introduced by an excessive number of parameters.

Gas Tracer Measurements in Beds of Large Particles

The design of a gas tracer experiment is mainly determined by the technique to be used for interpretation of experimental data. If a diffusion model is used then the boundary conditions of the appropriate diffusion equation are important. They determine the mode of the tracer injection and detection. In other words, the tracer experiments must be viewed as a part of the particular approach to the identification of gas flow characteristics.

In fluidized beds of large particles gas moves up through the interstices at a greater velocity than the velocity at which the bubbles rise, hence the bubbles provide a short-cut rather than a means of transportation to carry gas through the bed. Exploding and slow bubbles, Jovanovic, et al., 1980, are characteristic of a large particle fluidized bed and the effective bubble through flow is largely indicating that the rate of gas exchange within the bubble and emulsion phases is extremely high. The existing bubble emulsion models are adequate only for fine particle beds and would be unable to handle these differences.

Measuring the stimulus-response curve in coarse particle beds is difficult. These beds are usually shallow, 0.3 to 1.0 meters deep; the interstitial gas velocity can be as high as 3 m/s which means that an element of gas can leave the bed in less than 0.1 seconds. This requires instrumentation with a time resolution of a few milliseconds. Still, in order to investigate the behavior of a plume of volatiles released around the coal feed port in a fluidized bed

coal combustor, one must develop a stimulus-response curve. This can be accomplished by conducting impulse tracer injection experiments and measuring the mean residence time of the tracer gas which simulates released volatiles.

A diffusion type model provides a reasonable approach to modeling of the fluidized bed coal combustor; while the use of a two-region model of some kind appears to be very speculative at the moment.

With this in mind steady-state tracer experiments were conducted with the idea of using a diffusion model for interpretation of experimental data and impulse tracer injection experiments were used to measure the mean residence time of the volatiles in the bed.

The Mathematical Model

Figure 12 shows a representative tracer concentration profile across the bed at given height above the distributor plate and the tracer source (coal feed port). All points represent the time average of actual tracer signals. The concentration profile is a bell-shaped curve as expected.

Gifford (1959, 1960) proposed a "meandering" plume superimposed on an eddy diffusion concentration profile to describe overall exhaust stack plume dispersion. More recently, Jovanovic (1979) developed a similar model to describe tracer gas dispersion in a fluidized bed. The latter author presented experimental and analytical results for the case of a large source of tracer gas in a two-dimensional bed.

The meandering plume model for gas flow through a fluidized bed represents the convolution of two different mechanisms of gas motion which is schematically shown in Figure 13. One mechanism is that of turbulent and molecular diffusion of a gas volume, element dV , as it moves through the bed. The other mechanism

is that of lateral displacement of element dV by forces produced by bubble motion, bed internals, etc. This mechanism has been termed plume meander (Gifford, 1960) and it is assumed not to result in additional mixing of the gas from element dV with the surrounding gas, but rather it represents a lateral displacement of all of the gas in the vicinity of dV relative to some fixed point. In this study the lateral movement of the tracer gas is also enhanced by the plate mounted above the coal feed port. Therefore, the meandering dispersion coefficient, which quantifies the effect of meandering mechanism, includes as well the influence of the coal feed plate.

For this experimental work, the point of reference is taken as the location of the coal feed port, located at the center of the air distributor plate. A three-dimensional Cartesian coordinate system is assumed to originate at the coal feed port. The "z" direction is considered positive in the direction of the bulk gas movement. The "x" direction is parallel to a horizontal tube array which is immersed in the bed and the "y" direction is perpendicular to the axis of the tubes in the array.

The meandering effect, based on this coordinate system, is assumed to be described by a joint Gaussian probability density function with independence between x and y locations,

$$m(x,y,z) = \frac{1}{2\pi \sigma_{mx} \sigma_{my}} \exp\left[-\frac{1}{2}\left(\frac{x^2}{\sigma_{mx}^2} + \frac{y^2}{\sigma_{my}^2}\right)\right] \quad (1)$$

where

$$\sigma_{mx}^2 = \frac{2 D_{mx} z}{u} \rightarrow D_{mx} = \frac{\sigma_{mx}^2 u}{2z}$$

$$\sigma_{my}^2 = \frac{2 D_{my} z}{u} \rightarrow D_{my} = \frac{\sigma_{my}^2 u}{2z}$$

$m(x, y, z)$ represents the fraction of time that volume elements dV which originate at the tracer gas injector, will be found centered at (x, y, z) after a long observation time. Note that the above equation does not depend on z , except through the variances σ_{mx}^2 and σ_{my}^2 . Dispersion coefficients are, therefore, an indirect measure of the tracer spreading due to the meandering effect, i.e., the variances of this probability density function are directly proportional to dispersion coefficients.

For the experiments conducted with the plate above the coal feed port meandering dispersion coefficients in effect lump together the lateral displacement of the tracer gas caused by the plate itself, as well as the meandering motions inherent in this large particle fluidized bed.

At this time we have no reason to believe that meandering dispersion coefficients in "x" and "y" directions are different. Preliminary experiments and previous studies, Gosmeyer (1980), indicate that meandering dispersion coefficients, D_{mx} and D_{my} , are not statistically different, which simply means that the probability of the lateral displacement of the tracer gas at any location or radius r from the coal feed point is the same.

$$m(x, y, z) = \frac{u}{4\pi D_m z} \exp\left[-\frac{u}{4 D_m z} (x^2 + y^2)\right]. \quad (2)$$

However, if for some reason (skewed tube array, tilted and/or not centered coal feed port, misalignment of the gas jet of the coal feed excess air, etc.) gross and permanent circulation of particles is established (gulf streaming) the difference between meandering dispersion coefficients will be the measure of such gas flow distribution. The directional motion of particles could also be intentionally induced in fluidized beds of large particles, since the mixing of particles in such beds is rather poor (experiments toward better understanding of this phenomenon are currently underway at Oregon State University).

However, the effect of particle gulf streaming on gas distribution has not yet been studied. This may be very important in the case of fluidized bed coal combustors since the penetration of the volatiles into the freeboard may take place, which is not a desirable effect.

The turbulent and molecular dispersion along the meandering path is assumed to be described by Fick's second law as:

$$\nabla^2 D_i c = u \frac{\partial c}{\partial z} \quad (3)$$

which for the present case may be reduced to

$$D_x \frac{\partial^2 c}{\partial x^2} + D_y \frac{\partial^2 c}{\partial y^2} + D_z \frac{\partial^2 c}{\partial z^2} = u \frac{\partial c}{\partial z} \quad (4)$$

Jovanovic (1979) showed that the tracer data obtained in the experiments with a continuous tracer source does not give very reproducible values for the dispersion coefficients in the vertical direction, since the overall response is not sensitive to this parameter. This is just the consequence of the fact that the dispersion term in the "z" direction is much smaller than the convection term in the same direction

$$D_z \frac{\partial^2 c}{\partial z^2} \ll u \frac{\partial c}{\partial z} \quad (5)$$

In fact, except for the region very close to the injection point the calculated spread of tracer as a function of position is found to be about the same regardless of the value of the dispersion coefficient in the z direction. Several investigators came to the same conclusion. Rowe and Evans (1974) concluded that "the above model is insensitive to changes in the axial dispersion coefficient and depends mainly on the radial coefficient". Carberry (1976) also suggests that we can ignore the third term in Equation 4 since the dispersion term is small when compared to the convective term. Schugereel (1967) on

the otherhand suggests that all dispersion coefficients remain equal, arguing that dispersion is isotropic in gas fluidized beds. Gosmeyer (1980) came to the same conclusion after conducting tracer experiments in a scaled fluidized bed with immersed tubes. As noted earlier he also showed that the turbulent dispersion coefficient in "x" and "y" directions are not statistically different. Thus the decision is made to consider the fluidized bed as an isotropic media, i.e., to use the experimental data for the evaluation of dispersion coefficients D_{tx} , D_{ty} and D_{tz} for the case where all of them are equal.

The solution of Equation 4 for the continuous point source is first given by Wilson (1904),

for isotropic media

$$c(x,y,z) = \frac{\dot{M}}{4\pi D_t (x^2 + y^2 + z^2)^{3/2}} \exp \left\{ \frac{-u[(x^2 + y^2 + z^2)^{1/2} - z]}{2 D_t} \right\} \quad (6)$$

for anisotropic media

$$c(x,y,z) = \frac{\dot{M}}{4\pi [D_{tx} D_{ty} D_{tz} (\frac{x^2}{D_{tx}} + \frac{y^2}{D_{ty}} + \frac{z^2}{D_{tz}})^{1/2}]} \exp \left\{ \frac{u}{2 D_{tz}} \left[\frac{z}{D_{tz}} - (\frac{x^2}{D_{tx}} + \frac{y^2}{D_{ty}} + \frac{z^2}{D_{tz}})^{1/2} \right] \right\}. \quad (6a)$$

As mentioned before the time average concentration measured at point (x,y,z) is given by the convolution of the Equations 2 and 6.

$$\begin{aligned} \bar{c}(x,y,z) &= \frac{1}{T} \int_0^T c(x,y,z,t) dt = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} m(x',y',z) c(x-x',y-y',z) dx' dy' \\ &= \int_{-\infty}^{\infty} \frac{u}{4\pi z D_m} \exp \left[-\frac{u}{4z D_m} (x'^2 + y'^2) \right] \\ &\quad \times \frac{\dot{M}}{4\pi D_t [(x-x')^2 + (y-y')^2 + z^2]^{3/2}} \exp \left\{ \frac{-u}{2 D_t} [[(x-x')^2 + (y-y')^2 + z^2]^{1/2} - z] \right\} dx' dy' \end{aligned} \quad (7)$$

The time average mean of the square of the instantaneous concentrations is given by

$$\begin{aligned}\overline{c^2}(x,y,z) &= \frac{1}{T} \int_0^T c^2(x,y,z,t) dt = \int_{-\infty}^{\infty} m(x',y',z) c^2(x-x',y-y',z) dx' dy' \\ &= \overline{c^2}(x,y,z) = \int_{-\infty}^{\infty} \frac{u}{4\pi z D_m} \exp \left[-\frac{u}{4z D_m} (x'^2 + y'^2) \right] \\ &\quad \times \frac{\dot{M}^2}{16\pi^2 D_t^2 [(x-x')^2 + (y-y')^2 + z^2]} \exp \left\{ \frac{-u}{D_t} [[(x-x')^2 + (y-y')^2 + z^2]^{\frac{1}{2}} - z] \right\} dx' dy'\end{aligned}\quad (8)$$

Neither of these convolutions can be performed analytically. One way to alleviate the convolution integration problem of Equations 7 and 8 is to assume an approximate form for the diffusion concentration profile which can be integrated. A reasonable approximation (over the range of physical parameters encountered in the experimental apparatus) is given by

$$c(x,y,z) = \frac{\dot{M}}{4\pi D_t z} \exp \left[\frac{u}{4z D_t} (x^2 + y^2) \right] \quad (9)$$

Comparison of the Equations 6 and 9 shows good agreement for the actual experimental conditions. Using Equation 9 instead of Equation 6 we finally obtain for the convolutions indicated in Equations 7 and 8:

$$\overline{c}^*(x,y,z) = \exp \left[-\frac{u}{2z} \left(\frac{x^2 + y^2}{2D_t + 2D_m} \right) \right] \quad (10)$$

$$\overline{c^2}^*(x,y,z) = \exp \left[-\frac{u}{2z} \left(\frac{x^2 + y^2}{D_t + 2D_m} \right) \right] \quad (11)$$

where $\overline{c}^*$ and $\overline{c^2}^*$ are the normalized mean and mean square concentrations measured at points (x,y,z) with respect to $(0,0,z)$, i.e.;

$$\overline{c}^*(x,y,z) = \frac{\overline{c}(x,y,z)}{\overline{c}(0,0,z)} \quad \text{and} \quad \overline{c^2}^* = \frac{\overline{c^2}(x,y,z)}{\overline{c^2}(0,0,z)} \quad (11a)$$

The forms of the Equations 10 and 11 immediately suggest how the experimental data can be analyzed.

$$\ln \bar{c}^* = \frac{-u}{2z(2D_t + 2D_m)} (x^2 + y^2) \quad (12)$$

$$\ln \bar{c}^{2*} = \frac{-u}{2z(D_t + 2D_m)} (x^2 + y^2) \quad (13)$$

Since all of our data are taken in the plane (0,y,z) the above equations reduce to

$$\ln \bar{c}^* = \frac{-u}{2z(2D_t + 2D_m)} y^2; \quad (x = 0) \quad (14)$$

$$\ln \bar{c}^{2*} = \frac{-u}{2z(D_t + 2D_m)} y^2; \quad (x = 0) \quad (15)$$

Therefore, by plotting $\ln \bar{c}^*$ and $\ln \bar{c}^{2*}$ versus y^2 one should obtain straight lines whose slopes are proportional to $1/(2D_t + 2D_m)$ and $1/(D_t + 2D_m)$, respectively. (Note that u and z are parameters for a given set of runs.)

Figure 14 illustrates how one can obtain dispersion coefficients D_t and D_m . The overall observed dispersion coefficient is given by,

$$D_o = D_t + D_m \quad (16)$$

It is interesting to note that one can obtain the same final results, Equations 14 and 15, using different lines of reasoning.

The motion of the tracer gas in the bed can be viewed differently, especially for higher gas velocities, from the picture assumed so far.

Despite the fact that the point source of tracer gas emits tracer continuously, a violent motion of particles in the vicinity of the coal feed port can create conditions where an element of tracer gas is actually dispatched from

the tracer source in a small cloud. Such a cloud has an equal probability of going in any direction from the tracer source. Once in the fluidized bed, the cloud (an element of gas) meanders upward losing its identity due to the turbulent/molecular dispersion. The dispersion of the gas from the small clouds being randomly dispatched one after another (with equal probability in any direction from the tracer source) can be presented by the equation

$$c(x,y,z) = \frac{\dot{m}}{8(\pi D_t z/u)^{3/2}} \exp \left[\frac{-u}{4D_t z} (x^2 + y^2 + z^2) \right] \quad (17)$$

where $z = ut$ and $(x = 0, y = 0, z = 0)$ is the reference point.

The above equation is actually the solution of Equation 4 for the case of a moving point source in an infinite media as given by Carslaw and Jaeger (1952).

Now, following the same steps as in the case of Equations 7, 8, 10, 11, 12 and 13, we would come to the identical solution as in the case of Equations 14 and 15. Therefore, one can come to the same conclusions by replacing Equation 6 with an exact solution for the turbulent tracer dispersion, but only for the case where the dispersion mechanism is viewed as the tracer gas dispersion around an instantaneous moving point source in an infinite media.

Characteristic Fluidization Velocity

Often in the literature we find that the dispersion coefficients are calculated on the basis of superficial velocity, u_0 . This is a convenient approach since superficial velocity is a quantity easy to measure. However, u_0 is not a real characteristic fluidization velocity. Gas usually moves through the bed with velocities much different from u_0 . Use of superficial, interstitial or any other velocity can be accepted conventionally to represent a characteristic fluidization velocity to give "effective" dispersion coefficients, but only the true fluidization velocity can produce the true dispersion coefficients. Cau-

tion must be exercised when the results of two different studies are compared because different velocities could be used in evaluation of dispersion coefficients.

The characteristic fluidization velocity, " u ", appears as a parameter in many of the above equations. It is important to note that velocity " u " can be defined in a number of different ways. Rowe and Evans (1974) suggest that the excess gas velocity " $u_o - u_{mf}$ ", or as they call it "bubble flow", should be used in a diffusion equation which describes the motion of gas in fluidized beds. Their choice seems correct for tracer gas experiments performed in fine particle fluidized beds where the fast bubbles are the principal means of gas transport. However, in beds of large particles, bubbles are not vectors for the gas phase rather the interstitial gas moves faster than the bubbles. A small portion of gas rises up as very small bubble "clouds" while the rest of the gas uses the bubble as a short cut. Therefore, it would be incorrect to use an excess gas velocity as a characteristic fluidization velocity since only a very small portion of gas is transported by the bubbles themselves. A characteristic gas velocity defined as an interstitial velocity is a better alternative. This velocity is closer to the actual gas velocity inside the bed. Jovanovic (1979) uses this velocity as a characteristic fluidization velocity for the scope of his work.

However, the best solution to this problem is to measure the actual velocity of the gas through the bed. This information could be obtained from the mean residence time of tracer gas in the bed. Instrumentation with a fast time response for monitoring of the tracer concentration is necessary in order to obtain a reasonably good value for the mean residence time from the impulse response data.

Lehmann and Schugerl (1978) used mean residence time to define the characteristic fluidization velocity.

In this study a number of impulse-response experiments were performed in order to obtain mean residence times of the tracer gas, simulating volatiles release in the vicinity of the coal feed port.

CHAPTER 6

EXPERIMENTAL RESULTS

Characteristic Fluidization Velocity

Figures 15, 16 and 17 show the measured tracer velocity (characteristic fluidization velocity) values obtained from the impulse-response experiments. The upper curves in Figures 15 and 16 are for 20% transport air, the lower curves are for 10% transport air.

An impulse of tracer is introduced in the pipe which carries the transport air. Tracer concentration in the bed is monitored by two sampling probes. One of them is located immediately above the "coal feed port" while the second one is positioned on the surface of the slumped bed. During the run this probe is well below the surface of the bed due to the effect of the bed expansion. This upper probe can be moved laterally so that the tracer impulse-response can be measured at points which are not directly above the coal feed port.

The probes are calibrated by placing them at the same location in the bed, injecting a tracer impulse, and then measuring the time delay between the injection time and the detection time. This delay is a combination of the time required to transport the sample from the probe to the detector and the time required to detect the gas.

The tracer concentration is continuously recorded at a sampling rate of 100 samples per second, giving a time resolution of 10 milliseconds. The time necessary for the tracer impulse to pass between two probes is calculated as the difference between the recorded time of detection of the tracer impulse at the two sampling probes and their respective calibration times.

Usually 300 impulses are recorded before the mean residence time of the tracer gas is calculated. Therefore, each point on Figures 15 and 16 represents a mean of 300 measurements. Knowing the distance between the probes (32.6 cm) and the mean residence time of the tracer the interstitial velocity of the gas, i.e., the characteristic fluidization velocity can be calculated.

Figure 15 shows the characteristic fluidization velocities for the case where the coal feed port is not covered with a plate, for different superficial velocities (2, 3, 3.5, 4, 5 ft/s) and for two different percentages of coal transport air.

The same set of experiments are repeated for the case where the coal feed port is covered with a plate. The characteristic fluidization velocities for this case are shown in Figure 16.

The characteristic fluidization velocities for the cases of 10 and 20 percent of coal transport air are not significantly different due to the relatively large variances of the mean residence times obtained from most of the runs.

In addition, the characteristic fluidization velocities obtained from beds without the plates above the coal feed port are not significantly different from the corresponding velocities in beds with the plate in place.

Therefore, it is justified to consider all experimental points as data which come from one population. In that case, the mean tracer residence time could be calculated from all experimental points obtained for each of the superficial velocities. Figure 17 shows these points on the same graph.

All the curves representing the change of the characteristic fluidization velocity as a function of superficial velocity show identical trends. After an increase for lower superficial velocities, the curves begin leveling out.

at superficial velocities of 4 and 5 ft/s. This is in qualitative agreement with the findings of Jovanovic (1979) that the leveling of the curves was due to the large bed expansion in beds with tubes at high superficial velocities.

A caution must be exercised when this data is compared to data obtained in beds where all of the air is uniformly introduced in the bed through the distributor plate. The trend shown in Figures 15 and 16 suggest that the characteristic velocity would be 10 to 20 percent lower when all the air enters uniformly.

Dispersion Coefficients

In the steady-state gas tracer experiments tracer is continuously injected into a coal feed pipe and enters the bed with the transport air at the center of the distributor plate.

Three sample probes are used for continuously withdrawing a small quantity of gas at various points in the bed. Several ports located on the side of the bed provided a means of placing the probes at different axial positions. The axial positions were 22.2, 29.8, and 37.4 cm above the distributor plate. Once placed at a given axial position, each probe could move laterally to take the sample at any desired point. The exact lateral position was determined by measuring the length of the remainder of the probes outside the bed. The tracer concentration was measured at 10 lateral locations in order to trace the complete concentration profile at a given axial position.

The photo-ionization detectors are used for the continuous monitoring of the gas tracer concentration. The output analog signal (+10 to -10 volts) is sampled at a rate of 40 times a second for a period of 4 minutes and the

digitized data are stored on magnetic tapes.

The flow rate of tracer gas is adjusted before each run. All three probes are placed in the center of the bed directly above the tracer source. The optimal experimental conditions are obtained when the flow rate of tracer gas is increased up to a point where each channel gives a signal which is approximately 80% of the saturation reading. This requires that the amplification of the signal coming from the photo-ionization detectors is adjusted according to the distance of the sampling probes from the tracer source. The sample of gas taken by the probe closest to the injection line has larger amounts of tracer than the samples taken at the other locations. Consequently, the amplification of the signal on the first channel has to be reduced; otherwise, the level of the signal would exceed +10 volts which is the upper acceptable limit for the analog-to-digital converter. Typically, only one probe (usually the bottom one) show good resolution. Due to the very high values of the turbulent dispersion coefficients, the upper probes encounter the tracer so diluted with air that the resolution of the concentration profiles is rather poor.

After the adjustment, the system is left to operate for 15 minutes before any data are taken. The settings of the tracer gas flow rate and the signal amplifications are not changed until all data are collected for the given superficial velocity. When the superficial velocities are changed, the above procedure is repeated. The amount of tracer gas introduced into the system is not metered since the only requirement of the technique is that the flow rate must be constant during the experiment.

The level of the background signal is always determined before and after each series of runs and are subtracted from the raw data. The difference

between the two background readings is usually negligible because of the lengthy warm-up period.

Figures 18, 19 and 20 show dispersion coefficients as a function of superficial velocities and different amounts of coal transport air. These curves have the same typical shape which was previously observed by Jovanovic, et al. (1980) and Gossmeier (1980). For higher superficial velocities, the rate of increase of the dispersion coefficient is reduced almost to zero.

The most important feature of the curves is the ratio between the turbulent and meandering dispersion coefficients. While the turbulent dispersion coefficients increase with the superficial gas velocity. The meandering coefficients do not change appreciably. For higher superficial gas velocities, the ratio of the turbulent versus the meandering dispersion coefficients have values close to 4.

Another important feature of the data is shown in Figure 20. The turbulent dispersion coefficients obtained in the bed with 20 percent of the fluidization air introduced through the coal feed pipe are substantially higher (especially at higher superficial velocities) than the turbulent dispersion coefficients obtained in the bed where only 10 percent of coal transport air is introduced through the coal feed pipe. The values of the turbulent dispersion coefficient are in both cases much higher than the data reported by Gossmeier for no transport air (1980). It, therefore, appears that the introduction of relatively small portions of fluidization air through the coal feed pipe may be beneficial for the dispersion of volatiles released in the immediate vicinity of the coal feed port.

NOTATION

c	concentration of gas tracer (kg/m^3)
$\bar{c}$	time average concentration of gas tracer (kg/m^3)
$\bar{c}^*$	normalized time average concentration of gas tracer ($\bar{c}(y,z)/\bar{c}(0,z)$)
d_p	surface mean particle size (m)
D_o	overall dispersion coefficient (m^2/s)
D_m	meandering dispersion coefficient (m^2/s)
D_t	turbulent dispersion coefficient (m^2/s)
D, D_y, D_x	radial dispersion coefficient (m^2/s)
D_z	axial dispersion coefficient (m^2/s)
g	$= 9.81 \text{ (m/s}^2\text{)}$, acceleration of gravity
H	height of bed (m)
L	characteristic bed dimension (m)
L_2	radius of coal feed module (m)
$\dot{M}$	tracer source rate (moles/time)
t	time (s)
T	temperature ($^{\circ}\text{C}$)
u	characteristic fluidization velocity, u_o/ϵ (m/s)
u_o	superficial velocity (m/s)
x	characteristic distance (m)
y	horizontal distance from the gas tracer source (m)
z	vertical distance from the gas tracer

Greek Symbols

μ_f	viscosity of gas ($\text{kg/m}\cdot\text{s}$)
ρ_f, ρ_s	gas density, solid density (kg/m^3)
σ_m^2	$= \frac{D_m z}{u}$ mean square displacement of gas tracer due to the meandering of the fluid (m^2)
σ^2	$= \frac{D_z}{u}$ overall mean square displacement of gas tracer (m^2)

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Figure 1. Diagram of the main features of the bottom fed Atmospheric Fluidized Bed Combustor

Figure 2. Typical concentration profile of volatiles (A) and oxygen (B) in the vicinity of the plume.

Figure 3. Conversion of volatiles in a fluidized bed.

Figure 4. Temperature rise above the bed.

Figure 5. Criteria for distinguishing between slow and fast bubble fluidization regimes.

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Figure 7. Schematic of OSU's fluidized bed test facility.

Figure 8. Overall block diagram of gas-phase tracer system.

Figure 9. Schematic diagram of the photo-ionization detector.

Figure 10. Flow diagram of the photo-ionization electronic circuit.

Figure 11. Schematic of the photo-ionization amplifier circuit.

Figure 12. Typical time-averaged tracer concentration profile at a given height above the distributor plate.

Figure 13. Schematic of the mechanisms of gas motion through a fluidized bed.

Figure 14. Graphical representation of the numerical procedure employed in evaluation of the horizontal dispersion coefficient.

Figure 15. Characteristic fluidization velocity; bed without coal feed plate.

Figure 16. Characteristic fluidization velocity; bed with coal feed plate.

Figure 17. Characteristic fluidization velocity; combined data for bed with and without coal feed plate.

Figure 18. Overall D_o , Turbulent D_t , and Meandering D_m dispersion coefficients; 10% transport air, bed with coal feed plate.

Figure 19. Overall D_0 , Turbulent D_t , and Meandering D_m dispersion coefficients; 20% coal transport air, bed with coal feed plate.

Figure 20. Turbulent dispersion coefficients with 10% and 20% coal transport air.