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Computational analyses for modeling fluidized bed gasification processes

by

Nan Xie

A dissertation submitted to the graduate faculty in partial fulfillment of the requirements for the degree of

DOCTOR OF PHILOSOPHY

Major: Mechanical Engineering

Program of Study Committee: Francine Battaglia, Major Professor Rodney O. Fox Theodore J. Heindel Glenn R. Luecke Shankar Subramaniam

Iowa State University

Ames, Iowa

2007

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To my parents

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ABSTRACT

In-situ adaptive tabulation (ISAT) method, which can treat complex chemistry efficiently, has been implemented into a multiphase computational fluid dynamics (CFD) code. The numerical algorithm is improved to solve the chemical source term. Two-dimensional and three-dimensional non-isothermal detailed silane pyrolysis cases are used to test the features of ISAT, such as scale factors and multiple tables on multiple processors. ISAT can reduce the computational time for chemistry drastically. For example, a speed-up of 50 times is achieved with the non-isothermal three-dimensional case. Coal gasification with detailed chemical reactions is simulated to test the performance of ISAT with complex gas-solid reaction mechanisms. The speed-up of chemistry is around 16 times with 10% reduction in total CPU time for simulations. The elutriation phenomenon in fluidized beds is also studied in order to increase the efficiency of biomass reactors. In the simulations, fine and coarse particles are solved as two individual phases. Effects of gas velocity, and fine and coarse particle diameters are investigated for elutriation rate constants. The elutriation rate constants are proportional to approximately the 5.6th power of the gas velocity and decrease with increasing fine diameters. The diameters of coarse particles have little influence on elutriation rate constants. The simulation results are compared with experiments from the literature and performed at Iowa State University. Apart from the mainstream of this research, the effects on the use of coordinate systems and configurations to model fluidized bed reactors are tested. Three different fluidization regimes: bubbling, slugging and turbulent regimes are investigated. The results indicate that a two-dimensional Cartesian system can be used to successfully simulate and predict a bubbling regime. Caution must be exercised when using two-dimensional simulations for other fluidized regimes.

CHAPTER 1. INTRODUCTION

1.1 Background

Fluidized beds are found in many operations in chemical, petroleum, pharmaceutical, agricultural, biochemical, food, electronic, and power-generation industries. For example, combustion or gasification of coal is used in the power-generation industry. Fluid catalytic cracking (FCC) in chemical engineering industries employs one or more fluidized beds for a catalyst particles circulation system. Silicon for semiconductor industries is inexpensively produced from chemical vapor deposition (CVD) reactors.

The phenomenon of fluidization occurs when solid particles behave like a fluid state through suspension in a gas or liquid. Thus, a gas fluidized bed looks very much like a boiling liquid and in many ways exhibits liquid-like behavior. Gas-solid fluidized beds have advantages in comparison with other methods of gas-solid reactors (Kunii and Levenspiel, 1991). In fluidized beds, the rapid mixing of solids leads to nearly isothermal conditions throughout the bed, thereby, the whole bed of well-mixed solids resists rapid temperature changes. Heat and mass transfer rates between gas and particles are high compared to other methods. The smooth flow of particles allows continuous automatic operations and makes it possible to add or remove the vast quantities of heat in large reactors. Therefore, fluid-like behavior of solids with its rapid, easy transport and intimate gas contact in a fluidized bed are the most important characteristics for industrial applications.

Experiments are traditionally used in the design of multiphase reacting systems such as fluidized beds to improve efficiency of the process. However, a laboratory-scale process does not always exhibit the same flow behavior as a real reactor. It is often necessary to build expensive pilot-scale facilities in order to test the viability of fluidized beds. Computational fluid dynamics (CFD) provides an alternative method to analyze the gas-solid behavior. Usually, CFD calculations can give time-dependent information for pressure, temperature, composition and velocity distributions. With such information, the conditions in a reactor can be visualized and parametric studies can be conducted, thereby, assisting in the design process and avoiding the need for expensive pilot-scale tests. Barthod et al. (1999) have successfully improved the performance of a fluidized bed in the petroleum industry by using CFD.

CFD for reacting multiphase flows, however, is still not widely applied. One reason is because of the difficulty in describing multiphase systems. For example, there is still no agreement on detailed modeling of the interactions between the flowing gas and suspended particles (Wen and Yu, 1966; Gidaspow, 1994) or particle-particle interactions (Lun et al., 1984; Hrenya and Sinclair, 1997). The incorporation of chemical models in multiphase flow simulations is also a challenging task. Another issue is that multiphase flows are computationally expensive due to a wide range of length and time scales.

The entrainment of carbon particles from the freeboard is the main source of loss in reactor efficiency for fluidized bed gasifiers or combustors, especially for biomass reactors. Attrition and fragmentation for biomass are far more extensive since biomass usually has a higher moisture and volatile content, a more porous and fragile structure, a lower density and a higher intrinsic reactivity than coal. The efficiency of reactors is associated with the residence time of carbon fines in the bed. In addition, the elutriation of solid particles has to be known for designing gas cleaning equipment.

1.2 Problem Statement

The objective of this research is to advance the progress in simulating multiphase reacting flows using a government-developed numerical code. Multiphase Flow with Interphase eXchanges (MFIX) is a FORTRAN code developed by the National Energy Technology Laboratory (NETL) which models chemical reactions and heat transfer in dense and dilute fluid-solid flows. It can use multiple particle types, three-dimensional Cartesian or cylindrical coordinate systems and uniform or non-uniform grids. The continuity, momentum, energy and species equations are solved for gas and solid phases.

The first issue of this study is to improve the numerical algorithm to solve the chemical source term. In-situ adaptive tabulation (ISAT) (Pope, 1997) provides such a strategy and has a number of advantages compared to other tabulation schemes. For example, tables are generated during the calculations only for the regions in which chemical reactions happen. ISAT has been implemented into MFIX to test the feasibility of the time splitting method with a grid-based gas-solid code. It was the first study that demonstrated ISAT could be coupled to a finite-volume code. The test case was for the isothermal chemical vapor deposition of silane in a two-dimensional domain (Xie, 2002). There was only one chemical reaction with three gaseous species and two solid species.

In this research, first the MFIX-ISAT algorithm will be modified to improve ISAT performance. Then two-dimensional and three-dimensional non-isothermal detailed silane cases will be used to test the features of ISAT, such as scale factors and multiple tables on multiple processors. Based on the knowledge of the aforementioned tests, coal gasification with detailed chemical reactions will be simulated to test the performance of ISAT using complex gas-solid reaction mechanism.

The second task of this research is to study the elutriation of carbon in biomass reactors. In fluidized bed reactors, carbon comminution, such as fragmentation and attrition, occurs (Chirone et al., 1991). In combination with reactions, these phenomena control the reduction of feed carbon particles into fines of elutriable size, thereby, reactor efficiency. As the first step of this study, elutriation phenomenon will be simulated using two solid phases with the same density and different diameters. The simulation results will be validated with experiments from the literature and performed at Iowa State University.

1.3 Dissertation Organization

In Chapter 2, a survey of different modeling methods for multiphase flows is presented. The modeling methods include Eulerian-Eulerian modeling and Lagrangian-Eulerian modeling. Numerical methods for chemistry are then presented, which include methods like direct integration, reduction, and storage and retrieval. The governing equations and numerical methods for this research are described in detail in Chapter 3. The closure models for the equations such as solid phase stresses and interphase momentum transfer are presented. The implementation of chemical reactions using ISAT are presented in Chapter 4. Chapter 5 and 6 present the results of ISAT used in MFIX for silane decomposition and coal gasification. Chapter 7 shows the simulations of fluidized beds using different coordinate systems. The simulations of elutriation phenomenon using MFIX are compared with the experiments in Chapter 8. Chapter 9 discusses the conclusions and future work.

CHAPTER 2. LITERATURE REVIEW

Since there are many types of multiphase flows (gas-solid, gas-liquid, liquid-liquid, etc.) and several possible flow regimes (bubbly flow, slug flow, etc.) exist, general applicable models and methods are not available. Usually, two distinct numerical methods are used to simulate multiphase flow systems; namely the Lagrangian-Eulerian (LE) model or discrete element model (DEM) and the Eulerian-Eulerian (EE) model or two-fluid model (Sha, 1986). In addition, the chemical source term should be given close attention since a wide range of time scales appears in reactions. In this section, numerical methods which focus on gas-solid flows are introduced depending on the applications for multiphase flows with an overview of methods available for the chemical source term.

2.1 Survey of Methods for Solving Multiphase Flows

The early models developed for gas-solid flows were based on one-way coupling (Crowe, 1984). One-way coupling requires that the dispersed phase particles have no effect on the flow field of the continuous gas phase, but are controlled by the gas phase. Later on, numerical models incorporating two-way coupling were derived to describe the particle effects on the gas phase flow field. Four-way coupling methods are used for dense flows because the relative distance between particles is small enough so that particles collide. It is essential to examine the extent of coupling between gas and solid phases to select an appropriate modeling approach, especially for turbulent flows.

A classification of one-way, two-way and four-way coupling for gas-solid suspensions was proposed by Elghobashi (1991), based on the characteristic time scales and the volume fraction of particles as shown in Fig. 2.1. The particle relaxation time τ_{12}^x represents the entrainment



Figure 2.1 Classification of flow regimes for gas-solid flows by Elghobashi (1991).

of the particles by the gas phase, τ_1^t is the characteristic time scale of the large eddies in the continuous phase and ϵ_s is the volume fraction of solid phase. The assumption of one-way coupling is generally valid if the suspension is very dilute ($\epsilon_s < 10^{-6}$). Two-way coupling occurs when the particle volume fraction increases up to $\epsilon_s = 10^{-3}$. For a given volumetric fraction, turbulence is enhanced when the particle diameter is increased, whereas it is damped when the particle diameter is decreased. If particle volume fraction exceeds $\epsilon_s > 10^{-3}$, fourway coupling happens. In most fluidization applications, four-way coupling occurs, especially in the dense regions of the bed.

2.1.1 Eulerian-Eulerian Model

When different phases are mathematically described as interpenetrating continua, the Eulerian-Eulerian model is used to solve the equations for all phases. The continuity, momentum and energy equations with closure models are solved in all computational cells of the domain. Then the variables for different phases such as velocities and temperatures are known at every grid point. The continuity, momentum and energy equations for different phases in-teract through mass transfer, interaction forces representing the momentum transfer between

the phases, and interphase heat transfer. Examples of applications for the Eulerian-Eulerian model are circulating fluidized-bed combustors and fluid catalytic cracking risers. The model in this survey concentrates on gas-solid flows. However, the models and numerical methods are not necessarily restricted to gas-solid flows and most models can be applied to other kinds of mixtures.

Two sets of governing equations for the multiphase flow model originate from the works of Anderson and Jackson (1967) and Ishii (1975). Anderson and Jackson (1967) and Jackson (1997, 1998) derived the continuum equations of motion for gas-particle flow. They used a formal mathematical definition of local mean variables to translate the point variables for the fluid and the Newton's equation of motion for a single particle directly into continuum equations representing momentum balances for the fluid and solid phases. The point variables are averaged over large regions with respect to the particle diameter but the regions are considered small with respect to the characteristic dimension of the complete system. Ishii (1975) derived multiphase fluid-fluid governing equations in which both phases are averaged over a fixed volume; the volume is relatively large compared to the size of individual molecules.

The continuity equations have the same form in both descriptions by Anderson and Jackson (1967) and Ishii (1975). However, two differences appear in the momentum equations. First, the solid volume fraction multiplied by the gradient of the total gas-phase stress tensor ($\epsilon_s \nabla \cdot \bar{\tau}_g - \epsilon_s \nabla P$) is included in the solid-phase momentum equation by Anderson and Jackson (1967), whereas only the solid volume-fraction multiplied by the gradient of the pressure ($-\epsilon_s \nabla P$) is included by Ishii (1975). Second, in the gas-phase momentum balance of Ishii (1975), the pressure carries the gas volume fraction outside the gradient operator ($-\epsilon_g \nabla P$) whereas the shear stress carries the gas volume fraction inside the gradient operator ($\nabla \cdot \epsilon_g \bar{\tau}_g$). Both stresses are treated equally with respect to the gas volume fraction and the gradient operators ($-\epsilon_g \nabla P + \epsilon_g \nabla \cdot \bar{\tau}_g$) by Anderson and Jackson (1967). The results from both formulations for the governing equations are similar in terms of macroscopic flow behavior, but differ on a local scale such as individual bubbles or localized solid distributions (Wachem et al., 2001b). Closure of the solid-phase momentum equation requires a description for the solid-phase stress.

When the particle motion is dominated by collisional interactions, most of the researchers use the concept from gas kinetic theory for particles interaction. Other terms that need closure are drag force, gas-solid heat transfer and so on (Wachem et al., 2001b; Wachem and Almstedt, 2003).

The Eulerian-Eulerian approach is the practical approach for simulating large-scale particle flows. However, this approach needs sophisticated modeling to close the equations.

2.1.2 Lagrangian-Eulerian Model

The Lagrangian-Eulerian model is used in applications where one phase is continuous and the other phase is discrete and suspended. The continuous phase is solved using Eulerian methods on a computational grid, while the discrete suspended phase is solved using Lagrangian methods to track the trajectory of each particle. In one-way coupling, the continuous phase flow field can be obtained independent of the dispersed phase. Then the trajectories of particles can be calculated by the equations of motion. For two-way and four-way coupling, an iteration procedure is used since both phases interact with each other. The additional models (e.g., hard-sphere and soft-sphere models) for particle-particle interactions are needed for four-way coupling (Ranade, 2002). Examples of applications for the discrete particle model are blood flow and discrete particles in gas (Sha, 1986).

The motion of each particle is determined by the continuous phase and its neighboring particles. The equation for a particle which is subjected to Newton's equation of motion is

$$m\frac{\mathrm{d}\vec{v}}{\mathrm{d}t} = \sum \vec{F}$$

where \vec{F} is the force on the particle, m and \vec{v} are the mass and velocity of the particle respectively, and t is time. The continuous phase represents a generalization of the Naiver-Stokes equations.

The continuous phase exerts the traction force on the particles and influences their trajectories. Meanwhile, the particles are represented by the forces in the Stokes' equations of the continuous phase. Fogelson and Peskin (1986) studied three-dimensional incompressible flow with 500–1000 particles. Particle size, shape and deformability were prescribed, however, the interactions between the particles were neglected. The particle forces were spread to nearby grid points and the velocities of the continuous phase were interpolated from the grid to the particle points. The results came out well for sedimentation of particles under gravity and the computational time increased linearly with the number of particles. In discrete element models, the particles are influenced through a series of interactions with the continuous phase. The inertia effect, for example, was studied by Launay et al. (1998) in a homogeneous, isotropic turbulent flow. Ranade (2002) discussed the different drag coefficient correlations suitable for gas-solid flows.

Two types of approaches are possible for describing the collisions of particles: the hardsphere approach and soft-sphere approach. In the hard-sphere approach, collision between particles are assumed binary and instantaneous. Hoomans et al. (1996), Jie and Li (1999) and Wachem et al. (2001a) used this method to simulate the gas-solid flows. The physical motivation for the soft-sphere approach is that when two particles collide they actually deform. Tsuji et al. (1993) used the assumption of soft particles for interactions between particles to simulate gas-solid fluidization.

The Lagrangian-Eulerian model can track the trajectory of every particle. An advantage is the ability to easily vary the physical properties associated with individual particles such as size or density. Local physical phenomena related to the particle flow can be probed and can be used for validation and development of continuum models. However, the Lagrangian-Eulerian model needs a large memory requirement and long calculation time. Constitutive models are also required except when using direct numerical simulations for the continuous phase (Wachem et al., 2001b).

2.2 Numerical Modeling of the Chemical Source Term

Chemistry is an essential part in calculations of reactive flows and it forms the chemical source term in the governing equations. Fractional time stepping is widely used in reactive flows since it can avoid many costly matrix operations and allow the best numerical method to be used for each type of term (Oran and Boris, 2001). A consequence is that chemistry introduces a large number of coupled nonlinear ordinary differential equations. The nonlinear equations are notoriously stiff if the reactions cover a wide range of time scales. Different numerical methods have been developed to solve these equations (Yang and Pope, 1998), and are described next.

2.2.1 Direct Integration

Direct integration is a straightforward approach to determine the source term in which the reaction equation is integrated numerically. For a detailed reaction mechanism, direct integration is computationally expensive because the coupled differential equations for chemistry are stiff. For example methane combustion with a 16-species mechanism requires about 6 years of CPU time for 10⁹ times of direct integration on an SGI Indigo 2 workstation (Pope, 1997). Therefore, direct integration applied to detailed chemistry is prohibitive.

2.2.2 Reduction

Another approach developed to reduce the number of degrees of freedom in the description of chemistry are the reduced mechanism method (Peters, 1985) and the intrinsic lowdimensional manifold method (ILDM) (Maas and Pope, 1992). In reduced mechanism methods, a numerical analysis of a stoichiometric reaction is performed in order to determine the relative influence of any reaction in a large system of reactions. The maximum concentration level of all intermediate species then can be analyzed. For those species where the maximum mole fractions are sufficiently low and the diffusion coefficients are not too large, steady state assumptions are introduced. Then, for the reactions whose forward and backward rates are large compared to the other rate expressions, partial equilibrium is assumed. The algebraic expressions for the reaction rates are obtained by a partial equilibrium assumption. Peters (1985) reduced the chemical reactions for hydrocarbon flames from eighteen to four using reduced mechanism methods.

Some drawbacks exist for the reduced mechanism methods. These methods require a

considerable amount of human time and experience to develop the reduced scheme for each reaction system, or, even for the same reaction system under different conditions. Assumptions of steady state and partial equilibrium are not always accurate.

The ILDM is based on the intrinsic study of underlying time scales of chemical reactions. The fast time scales of chemical reactions are decoupled by assuming local equilibrium. The method reduces the number of dimensions of reactions to the non-linear manifold, defined by the slowest chemical time scales. The number of slow chemical scales is very small, for example, one to three for combustion systems. The $CO/H_2/air$ system was used to test the ILDM (Maas and Pope, 1992). However, it is not straightforward to find and parameterize the manifold for a detailed reactions scheme because the low-dimensional manifold is non-linear.

2.2.3 Storage and Retrieval

In storage and retrieval methods, the chemical source terms are stored in multidimensional databases which are based on direct integration of either a full mechanism or a reduced mechanism. The databases can be generated during the simulations or ahead of time.

Pre-computed look-up tabulation is one type of storage and retrieval method (Fox, 2003). Since the chemical source term is computationally expensive, a pre-computed chemical lookup table is calculated and stored for a set of representative initial conditions in composition space. The simplest method is to place these points on a multidimensional grid in composition space. Then numerical interpolation is used to find the values based on the neighboring points, replacing direct integration in the simulations. Taing et al. (1993) used pre-computed look-up tabulation in Monte Carlo calculations of the joint velocity-composition probability density function for pilot-stabilized turbulent jet nonpremixed flames of H_2/CO_2 fuel mixtures. The multidimensional look-up tables are generated using a three-step chemical kinetic scheme, reduced systematically from a more detailed mechanism to represent chemical reactions. These tables were used for density, and other dependent properties such as temperature and composition. Taing et al. (1993) demonstrated that the use of these tables in calculations was computationally efficient. Tabulation methods have drawbacks, however. The whole accessible region of the composition space must be tabulated since it is not known which regions are needed before simulations. Thus, the required table is very large, especially when the number of reaction dimensions is high. When the dimension of the table is high, the work to retrieve information from the table is not trivial. Therefore, the application of pre-computed look-up tables will be limited by the complexity of reactions. The direct application to detailed kinetics is not feasible.

In-situ adaptive tabulation (ISAT) is a method which overcomes many of the difficulties of look-up tables and this method is explained in Chapter 4 in detail. Another example of storage and retrieval is the repro-modeling approach (Turnyi, 1994). In the repro-modeling approach, information for reaction rates is extracted from detailed chemical calculations and stored in the form of high-order multivariate polynomials.

According to Pope (1997), criteria for the storage and retrieval techniques are

- 1. the CPU time required to create the store,
- 2. the memory required for the store,
- 3. inaccuracies in the retrieved table (e.g. interpolation error),
- 4. the CPU time required to retrieve the table.

The retrieval time may be three orders of magnitude smaller than the time to perform a direct integration. Therefore, if the number of retrievals is over 10^9 , the time for creating the table is negligible.

CHAPTER 3. THEORETICAL FORMULATION

In this chapter, a mathematical model for multiphase flow based on hydrodynamic theory of fluidization is presented. The models necessary to close the system of equations are introduced. Finite volume schemes are used to discretize the governing equations. The SIMPLE algorithm is modified for multiphase flow to solve the algebraic equations.

3.1 Hydrodynamic Theory

The hydrodynamic model for multiphase flow assumes that different phases behave as interpenetrating continua and the instantaneous variables are averaged over a region which is larger than the particle spacing but much smaller than the flow domain. Multiple types of particles should be considered to describe phenomena such as particle segregation and elutriation. Each particle type has a unique diameter with identical material properties (Syamlal et al., 1993).

3.1.1 Governing Equations

The field variables, called phasic volume fractions, are used to determine the fraction of the average volume occupied by each phase. The sum of all of the volume fractions, both for the gas phase and the m^{th} solid phases, where m denotes a unique solid phase, must equal unity

$$\epsilon_{\rm g} + \sum_{m=1}^{\rm M} \epsilon_{\rm sm} = 1$$

The subscript g indicates the gas phase and s indicates solid phases with the total number of solid phase M. The gas phasic volume fraction is also called the void fraction. The effective

densities of the gas phase $\rho_{\rm g}'$ and solid phases $\rho_{\rm sm}'$ respectively, are

$$ho_{\rm g}' = \epsilon_{\rm g}
ho_{\rm g}$$

 $ho_{\rm sm}' = \epsilon_{\rm sm}
ho_{\rm sm}$

The continuity equations for the gas phase and the solid phase m, respectively, are

$$\frac{\partial}{\partial t} (\epsilon_{\rm g} \rho_{\rm g}) + \nabla \cdot (\epsilon_{\rm g} \rho_{\rm g} \vec{v}_{\rm g}) = \sum_{n=1}^{N_{\rm g}} R_{\rm gn}$$
(3.1)

$$\frac{\partial}{\partial t}(\epsilon_{\rm sm}\rho_{\rm sm}) + \nabla \cdot (\epsilon_{\rm sm}\rho_{\rm sm}\vec{v}_{\rm sm}) = \sum_{n=1}^{N_{\rm sm}} R_{\rm smn}$$
(3.2)

The first and second terms on the left sides of Eqs. (3.1) and (3.2) are the rate of mass generation per unit volume and the net rate of convective mass flux. The terms of R_{gn} and R_{smn} on the right sides account for interphase mass transfer due to chemical reactions or physical processes.

The momentum equations for the gas phase and the solid phase m have the form

$$\frac{\partial}{\partial t} (\epsilon_{\rm g} \rho_{\rm g} \vec{v}_{\rm g}) + \nabla \cdot (\epsilon_{\rm g} \rho_{\rm g} \vec{v}_{\rm g} \vec{v}_{\rm g}) = \nabla \cdot \overline{\overline{S}}_{\rm g} + \epsilon_{\rm g} \rho_{\rm g} \vec{g} - \sum_{m=1}^{M} \vec{I}_{\rm gm} + \vec{f}_{\rm g}$$
(3.3)

$$\frac{\partial}{\partial t}(\epsilon_{\rm sm}\rho_{\rm sm}\vec{v}_{\rm sm}) + \nabla \cdot (\epsilon_{\rm sm}\rho_{\rm sm}\vec{v}_{\rm sm}\vec{v}_{\rm sm}) = \nabla \cdot \overline{\overline{S}}_{\rm sm} + \epsilon_{\rm sm}\rho_{\rm sm}\vec{g} + \vec{I}_{\rm gm} - \sum_{\substack{l=1\\l\neq m}}^{\rm M}\vec{I}_{ml} \qquad (3.4)$$

The first terms on the left sides of Eqs. (3.3) and (3.4) are the net rate of momentum generation and the second terms are the net rate of momentum transfer by convection. Equations (3.3) and (3.4) include the gas phase and solid phase stress tensors \overline{S}_g and \overline{S}_{sm} , the body forces due to gravity \vec{g} , the flow resistance force \vec{f}_g due to internal porous surfaces, the interaction force \vec{I}_{gm} accounting for the momentum transfer between the gas phase and the m^{th} solid phase, and the interaction force \vec{I}_{lm} between the m^{th} and l^{th} solid phases. The internal energy balances for the gas and solid phase m are

$$\epsilon_{\rm g}\rho_{\rm g}C_{\rm pg}\left(\frac{\partial T_{\rm g}}{\partial t} + \vec{v}_{\rm g}\cdot\nabla T_{\rm g}\right) = -\nabla\cdot\vec{q}_{\rm g} - \sum_{m=1}^{\rm M}H_{\rm gm} - \triangle H_{\rm rg} + H_{\rm wall}(T_{\rm wall} - T_{\rm g}) \tag{3.5}$$

$$\epsilon_{\rm sm}\rho_{\rm sm}C_{\rm psm}\left(\frac{\partial T_{\rm sm}}{\partial t} + \vec{v}_{\rm sm} \cdot \nabla T_{\rm sm}\right) = -\nabla \cdot \vec{q}_{\rm sm} + H_{\rm gm} - \triangle H_{\rm rsm}$$
(3.6)

with temperature T and constant pressure specific heats C_p . Equations (3.5) and (3.6) include the conductive heat flux \vec{q}_g and \vec{q}_{sm} , the gas-solid interphase heat transfer H_{gm} , and the heats of reaction ΔH_{rg} and ΔH_{rsm} . The last term in Eqn. (3.5) is the heat loss to the wall. Heat transfer between different solid phases is negligible due to rapid mixing and uniform temperature distribution in the dense bed.

The gas and solid phases are formulated to represent an arbitrary number of chemical species, $N_{\rm g}$ and $N_{\rm sm}$. The corresponding conservation of species for the gas and solid phases are

$$\frac{\partial}{\partial t} (\epsilon_{\rm g} \rho_{\rm g} X_{\rm gn}) + \nabla \cdot (\epsilon_{\rm g} \rho_{\rm g} X_{\rm gn} \vec{v}_{\rm g}) = \nabla \cdot D_{\rm gn} \nabla X_{\rm gn} + R_{\rm gn}$$
(3.7)

$$\frac{\partial}{\partial t}(\epsilon_{\rm sm}\rho_{\rm sm}X_{\rm smn}) + \nabla \cdot (\epsilon_{\rm sm}\rho_{\rm sm}X_{\rm smn}\vec{v}_{\rm sm}) = \nabla \cdot D_{\rm smn}\nabla X_{\rm smn} + R_{\rm smn}$$
(3.8)

where X is the mass fraction and D is the diffusion coefficient. Equations (3.7) and (3.8) include accumulation, convection and the rate of reaction but neglect diffusive fluxes.

The chemical source terms are closed by reaction kinetic expressions in the species balance equations. It should be noted that the chemical source terms involve a wide range of time scales which will make the equations stiff. Therefore, efficient algorithms must be used to treat these terms.

3.1.2 Constitutive Relations

The complete formulation requires that the governing equations be closed. A wide range of constitutive relations can be found by using different approaches, from empirical information to kinetic theory. Most differences between multiphase theories originate from closure assumptions. The closure models used in this research are introduced next.

3.1.2.1 Gas-Phase Equation of State

The equation of state for the gas phase assumes ideal gas behavior,

$$\rho_{\rm g} = \frac{P_{\rm g} M_{\rm w}}{RT_{\rm g}}$$

where $P_{\rm g}$ is the gas pressure, $M_{\rm w}$ is the molecular weight and R is the universal gas constant. For an incompressible gas the density is assumed to be constant.

3.1.2.2 Gas-Solid Momentum Transfer

From previous studies on the dynamics of a single particle in a gas, three important mechanisms for the gas-solid interaction force \vec{I}_{gm} are considered: drag force, caused by velocity differences between gas-solid phases; buoyancy, caused by the gas pressure gradient; and momentum transfer due to mass transfer between phases. Other forces, such as virtual mass effect caused by relative acceleration between phases, Magnus force caused by particle spin and so on, are not considered. Thus, the gas-solid momentum interaction force becomes

$$\vec{I}_{gm} = -\epsilon_{sm} \nabla P_{g} - F_{gm} (\vec{v}_{sm} - \vec{v}_{g}) + R_{0m} [\zeta_{0m} \vec{v}_{sm} + \bar{\zeta}_{0m} \vec{v}_{g}]$$
(3.9)

where R_{0m} is the mass transfer from the gas phase to solid phase m, and

$$\zeta_{0m} = \begin{cases} 1 & \text{for } R_{0m} < 0 \\ \\ 0 & \text{for } R_{0m} > 0 \end{cases}$$

for $\overline{\zeta}_{0m} = 1 - \zeta_{0m}$,

The gas-solid drag formulation for converting terminal velocity correlations to drag corre-

lations (Syamlal and O'Brien, 1987) is

$$F_{\rm gm} = \frac{3\epsilon_{\rm sm}\epsilon_{\rm g}\rho_{\rm g}}{4V_{rm}^2 d_{\rm pm}} C_{\rm DS} \left(\frac{Re_m}{V_{rm}}\right) |\vec{v}_{\rm sm} - \vec{v}_{\rm g}|$$
(3.10)

where d_{pm} is the diameter and V_{rm} is the terminal velocity correlation for the m^{th} solid phase.

$$V_{rm} = 0.5 \left[A - 0.06Re_m + \sqrt{(0.06Re_m)^2 + 0.12Re_m(2B - A) + A^2} \right]$$
(3.11)

where A and B are the functions of void fraction. The Reynolds number of the m^{th} solid phase is

$$Re_m = \frac{d_{\rm pm} |\vec{v}_{\rm sm} - \vec{v}_{\rm g}|\rho_{\rm g}}{\mu_{\rm g}} \tag{3.12}$$

The single sphere drag function (Dalla Valla, 1948) has the form

$$C_{\rm DS}\left(\frac{Re_m}{V_{rm}}\right) = \left(0.63 + \frac{4.8}{\sqrt{Re_m/V_{rm}}}\right)^2 \tag{3.13}$$

Equations (3.11), (3.12) and (3.13) are coupled to (3.10) for the complete description of the gas drag coefficient.

3.1.2.3 Solid-Solid Momentum Transfer

The drag between the phases due to velocity differences is assumed as the major effect in the solid-solid momentum transfer. Thus, the momentum transfer I_{ml} between solids is represented as

$$\vec{I}_{ml} = -F_{sml}(\vec{v}_{sl} - \vec{v}_{sm}) + R_{ml}[\zeta_{ml}\vec{v}_{sl} + \bar{\zeta}_{ml}\vec{v}_{sm}]$$
(3.14)

where R_{ml} is the mass transfer from solid phase m to solid phase l,

$$\zeta_{ml} = \begin{cases} 1 & \text{for } R_{ml} < 0 \\ 0 & \text{for } R_{ml} > 0 \end{cases}$$

and $\bar{\zeta}_{ml} = 1 - \zeta_{ml}$,

The drag coefficient F_{sml} is derived by using a simplified version of kinetic theory (Syamlal, 1987a)

$$F_{\rm sml} = \frac{3(1+e_{lm})(\pi/2 + C_{\rm flm}\pi^2/8)\epsilon_{\rm sl}\rho_{\rm sl}\epsilon_{\rm sm}\rho_{\rm sm}(d_{\rm pl} + d_{\rm pm})^2 g_{0_{lm}}|\vec{v}_{\rm sl} - \vec{v}_{\rm sm}|}{2\pi(\rho_{\rm sl}d_{\rm pl}^3 + \rho_{\rm sm}d_{\rm pm}^3)}$$
(3.15)

where e_{lm} and C_{flm} are the coefficient of restitution and coefficient of friction, between the l^{th} and m^{th} solid-phase particles, and $g_{0_{lm}}$ is the radial distribution function at contact for a mixture of hard spheres.

3.1.2.4 Gas Phase Stress Tensor

The gas phase stress tensor has two parts: the pressure contribution and the Newtonian viscous stress tensor, which is given by

$$\overline{\overline{S}}_{g} = -P_{g}\overline{\overline{I}} + \overline{\overline{\tau}}_{g} \tag{3.16}$$

The viscous stress tensor, $\overline{\overline{\tau}}_{g}$, is

$$\overline{\overline{\tau}}_{g} = 2\epsilon_{g}\mu_{g}\overline{\overline{D}}_{g} + \epsilon_{g}\lambda_{g}\mathrm{tr}(\overline{\overline{D}}_{g})\overline{\overline{I}}$$
(3.17)

where $\overline{\overline{I}}$ is the identity tensor and $\overline{\overline{D}}_{g}$ is the strain rate tensor for the gas phase

$$\overline{\overline{D}}_{\mathrm{g}} = \frac{1}{2} \left[\nabla \vec{v}_{\mathrm{g}} + (\nabla \vec{v}_{\mathrm{g}})^T \right]$$

3.1.2.5 Solid Phase Stress Tensor

In the solid phase stress tensor term, a solid pressure is calculated to ensure that the volume fraction does not exceed the packed-bed volume fraction. This can be accomplished by treating the particles as an incompressible fluid at a critical void fraction and the pressure is set to zero when the void fraction becomes greater than the packed-bed void fraction (Syamlal and O'Brien, 1988).

Granular flow theory is used to describe the solid phase stresses in detail. There are two distinct flow regimes in granular flow: a viscous or rapidly shearing regime in which stresses arise due to collisional or translational momentum transfer, and a plastic or slowly shearing regime in which stresses arise due to Coulomb friction between grains in close contact. Two different approaches are used for these regimes:

$$\overline{\overline{S}}_{\mathrm{s}m} = \begin{cases} -P_{\mathrm{s}m}^{\mathrm{p}} \overline{\overline{I}} + \overline{\overline{\tau}}_{\mathrm{s}m}^{\mathrm{p}} & \text{if } \epsilon_{\mathrm{g}} \leq \epsilon_{\mathrm{g}}^{*} \\ \\ -P_{\mathrm{s}m}^{\mathrm{v}} \overline{\overline{I}} + \overline{\overline{\tau}}_{\mathrm{s}m}^{\mathrm{v}} & \text{if } \epsilon_{\mathrm{g}} > \epsilon_{\mathrm{g}}^{*} \end{cases} \end{cases}$$

where P_{sm}^{p} and $\overline{\overline{\tau}}_{sm}^{p}$ are the pressure and the viscous stress in the m^{th} solid phase for the plastic regime, P_{sm}^{v} and $\overline{\overline{\tau}}_{sm}^{v}$ are the pressure and the viscous stress for the viscous regime, and ϵ_{g}^{*} is a critical packing number, usually set to the void fraction at minimum fluidization.

The granular stress equation based on kinetic theory (Syamlal, 1987b) is applied to the viscous regime. The granular pressure and stresses are given by

$$P_{\rm sm}^{\rm v} = K_{1m} \epsilon_{\rm sm}^2 \Theta_m \tag{3.18}$$

$$\overline{\overline{\tau}}_{\mathrm{s}m}^{\mathrm{v}} = 2\mu_{\mathrm{s}m}^{\mathrm{v}}\overline{\overline{D}}_{\mathrm{s}m} + \lambda_{\mathrm{s}m}^{\mathrm{v}}\mathrm{tr}(\overline{\overline{D}}_{\mathrm{s}m})\overline{\overline{I}}$$
(3.19)

where Θ_m is the granular temperature and λ_{sm}^v is the second coefficient of viscosity,

$$\lambda_{\mathrm{s}m}^{\mathrm{v}} = K_{2m} \epsilon_{\mathrm{s}m} \sqrt{\Theta_m}$$

The shear viscosity factor $\mu_{\mathrm sm}^{\mathrm v}$ is

$$\mu_{\mathrm{s}m}^{\mathrm{v}} = K_{3m} \epsilon_{\mathrm{s}m} \sqrt{\Theta_m}$$

The strain rate tensor, $\overline{\overline{D}}_{sm}$, is given by

$$\overline{\overline{D}}_{\mathrm{s}m} = \frac{1}{2} \left[\nabla \vec{v}_{\mathrm{s}m} + (\nabla \vec{v}_{\mathrm{s}m})^T \right]$$
and K_{1m} , K_{2m} and K_{3m} are constants.

In the plastic flow regime, frictional flow theory (Jenike, 1987) provides an arbitrary function that allows a certain amount of compressibility in the solid phase where

$$P_{\rm sm}^{\rm p} = \epsilon_{\rm sm} P^* \tag{3.20}$$

and P^* is expressed by an empirical power law

$$P^* = A(\epsilon_{\rm g}^* - \epsilon_{\rm g})^n$$

for values of $A = 10^{25}$ and n = 10.

The solid stress tensor is calculated for solid phase m using the formulation (Schaeffer, 1987)

$$\overline{\overline{\tau}}_{\rm sm}^{\rm p} = 2\mu_{\rm sm}^p \overline{\overline{D}}_{\rm sm} \tag{3.21}$$

3.1.2.6 Gas-Solid Heat Transfer

The heat transfer between the gas and solid phases is a function of a temperature difference

$$H_{\rm gm} = -\gamma_{\rm gm} (T_{\rm sm} - T_{\rm g}) \tag{3.22}$$

where γ_{gm} is the heat transfer coefficient between the gas phase and the m^{th} solid phase. The heat transfer coefficient γ_{gm} (Bird et al., 1960) is corrected using the coefficient γ_{gm}^0 by adding the influence of the interphase mass transfer,

$$\gamma_{\rm gm} = \frac{C_{\rm pg} R_{0m}}{e^{(C_{\rm pg} R_{0m} / \gamma_{\rm gm}^0)} - 1}$$

and the coefficient γ_{gm}^0 is

$$\gamma_{\rm gm}^0 = \frac{6k_{\rm g}\epsilon_{\rm sm}Nu_m}{d_{\rm pm}^2}$$

The correlation is applicable for a porosity range of 0.35–1.0 and a Reynolds number up to

 10^5 . For the work herein, the cases studied fall within the ranges of the correlation.

3.1.2.7 Conductive Heat Flux in Gas and Solid Phase

The conductive heat fluxes for both the gas and solid phases are described according to Fourier's law

$$\vec{q}_{\rm g} = -\epsilon_{\rm g} k_{\rm g} \nabla T_{\rm g} \tag{3.23}$$

$$\vec{q}_{\rm sm} = -\epsilon_{\rm sm} k_{\rm sm} \nabla T_{\rm sm} \tag{3.24}$$

where $k_{\rm g}$ and $k_{\rm sm}$ are the thermal conductivity for gas and particle phases, respectively. The particle thermal conductivity (Syamlal and Gidaspow, 1985) has the form

$$\frac{k_{\rm sm}}{k_{\rm g}} = \frac{\left[\Phi_k R_{km} + (1 - \Phi_k)\lambda_{rm}\right]}{\sqrt{1 - \epsilon_{\rm g}}}$$

where Φ_k is the contact area function with the value 7.26×10^{-3} and R_{km} and λ_{rm} are constants.

3.1.2.8 Heat of Reaction

The heat of reaction accounts for the difference in temperatures between phases by partitioning reaction processes between each phase. Partitioning of the heat is arbitrary since the averaging required to derive the hydrodynamic equations does not contain any information regarding the gas-solid interface. The actual chemical reactions occur in an interface region, however, the partitioning of the heats of reaction must be based on physical arguments. For example, to partition the heat of the coal combustion reaction $C + O_2 \rightarrow CO_2$, the heat of reaction for the step $C + \frac{1}{2}O_2 \rightarrow CO$ is assigned to the solid phase ($\triangle H_{\rm rs}$ in Eqn. (3.6)) and the heat of reaction for the step $CO + \frac{1}{2}O_2 \rightarrow CO_2$ is assigned to the gas phase ($\triangle H_{\rm rg}$ in Eqn. (3.5)).

3.1.2.9 Granular Energy Equations

The constitutive relation for granular flow is based on kinetic theory of smooth, slightly inelastic, spherical particles. The relationship contains the granular temperature Θ_m which is proportional to the granular energy of the continuum. The granular temperature is introduced as a measure of the particle velocity fluctuations

$$\frac{3}{2}\Theta_m = \frac{1}{2} \langle \vec{C}_m^2 \rangle$$

where \vec{C}_m is the fluctuating component of the instantaneous velocity \vec{c}_m of the m^{th} solid phase

$$\vec{c}_m = \vec{v}_{\rm sm} + \vec{C}_m$$

The transport of granular energy is

$$\frac{3}{2} \left[\frac{\partial (\epsilon_{\rm sm} \rho_{\rm sm} \Theta_m)}{\partial t} + \nabla \cdot (\epsilon_{\rm sm} \rho_{\rm sm} \Theta_m) \vec{v}_{\rm sm} \right] = \overline{\overline{S}}_{\rm sm} : \nabla \vec{v}_{\rm sm} - \nabla \cdot \vec{q}_{\Theta_m} - \gamma_{\Theta_m} + \Phi_{\rm gm} + \sum_{l=1, l \neq m}^{\rm M} \Phi_{lm}$$
(3.25)

where γ_{Θ_m} is the rate of granular energy dissipation due to inelastic collisions of the form (Lun et al., 1984)

$$\gamma_{\Theta_m} = K_{4m} \epsilon_{\mathrm{sm}}^2 \Theta_m^{\frac{3}{2}}$$

and \vec{q}_{Θ_m} is the diffusive flux of granular energy

$$\vec{q}_{\Theta_m} = -k_{\Theta_m} \nabla \Theta_m$$

The term Φ_{gm} is the transfer of granular energy between the gas phase and the m^{th} solid phase, and Φ_{lm} accounts for the transfer of granular energy between the m^{th} and l^{th} solid phases. The formulation for granular energy transfer (Ding and Gidaspow, 1990) is

$$\Phi_{\rm gm} = -3F_{\rm gm}\Theta_m$$

and Φ_{lm} is ignored by setting $\Phi_{lm} = 0$.

The granular temperature Θ_m is calculated using an algebraic expression by assuming that the granular energy is dissipated locally and the convection and diffusion contributions are neglected. The simplification is a reasonable assumption in the case of fluidized-bed modeling and reduces the computational effort by about 20% according to Wachem et al. (2001b). The algebraic granular energy equation used is

$$\Theta_m = \left\{ \frac{-K_{1m}\epsilon_{\rm sm} {\rm tr}(\overline{\overline{D}}_{\rm sm}) + \sqrt{K_{1m}^2 {\rm tr}^2(\overline{\overline{D}}_{\rm sm})\epsilon_{\rm sm}^2 + 4K_{4m}\epsilon_{\rm sm}[K_{2m} {\rm tr}^2(\overline{\overline{D}}_{\rm sm}) + 2K_{3m} {\rm tr}(\overline{\overline{D}}_{\rm sm}^2)]}{2\epsilon_{\rm sm}K_{4m}} \right\}^2$$

$$(3.26)$$

3.1.3 Initial and Boundary Conditions

3.1.3.1 Initial Conditions

The initial values of all field variables must be specified for the entire computational domain. However, the initial conditions are usually not of interest, and the solution is governed by the boundary conditions. Therefore, the initial conditions merely affect the initial convergence of the equations.

3.1.3.2 Boundary Conditions

Two types of inflow boundary conditions are possible, constant velocity or constant mass flux. Specifying constant pressure is the most common condition for the fluid outflow boundary. At impermeable walls, the normal velocities are set to zero. The condition for the tangential components is specified either as a non-slip or as a free-slip condition. The fixed temperature and specified heat flux can be used as thermal boundary conditions. For species equations, either specified species concentration or species diffusion flux can be used.

3.2 Numerical Formulation

A finite volume approach is used for the numerical discretization of the equations for multiphase flows. The approach has an advantage to ensure the conservation laws for mass, momentum and energy. The conservative property of finite volume methods is more attractive, especially if a fine grid is too computationally expensive. Thus, even for coarse grids, the solutions hold the integral balances (Patankar, 1980).

3.2.1 A Staggered Grid

A staggered grid is used for discretizing the partial differential equations. Scalars such as pressure, void fraction, temperature, and mass fraction are stored at the cell centers and the velocity components are stored at the cell surfaces. If all variables such as velocity and pressure are stored on the same grid points, the pressures for two alternate grid points appear in the momentum equations. Thus the zigzag pressure field for a one-dimensional domain and checkerboard pressure field for a two-dimensional domain develop as an acceptable solution. More complex results exist for three-dimensional case and a similar situation appears for the velocity components from continuity equations (Patankar, 1980). If a staggered grid is used, unphysical pressure and velocity fields will not occur since two neighboring grid points appear in the momentum equations.

3.2.2 Discretization

3.2.2.1 Discretization of the Governing Equations

The discretization of the governing equations is discussed using the transport equation for a scalar variable Φ

$$\frac{\partial}{\partial t}(\epsilon_m \rho_m \Phi) + \frac{\partial}{\partial x_i}(\epsilon_m \rho_m v_{mi} \Phi) = \frac{\partial}{\partial x_i} \left(\Gamma_\Phi \frac{\partial \Phi}{\partial x_i}\right) + R_\Phi \tag{3.27}$$

where m = 0 denotes the gas phase, m = 1 to M denotes the solid phases, Γ_{Φ} is the diffusion coefficient for Φ and R_{Φ} is the source term.

Equation (3.27) has all the features of the equations for multiphase flow except the interphase transfer term. The interphase transfer term is important in the multiphase flow equations and is discussed in §3.2.2.2. The equation is integrated over a control volume. Figure 3.1 shows a computational cell and the nomenclature of discretization (Syamlal, 1994). The point P is



Figure 3.1 Computational cell for the scalar equations.

at the center of the control volume and its neighboring points are referred to as E, W, N, S, T, B. The half-node positions are referred to as e, w, n, s, t, b.

A one-dimensional scalar equation is used as an example for discretization. Convectiondiffusion terms have the following form in the x-direction

$$\rho u \frac{\partial \Phi}{\partial x} - \frac{\partial}{\partial x} \left(\Gamma \frac{\partial \Phi}{\partial x} \right) \tag{3.28}$$

Equation (3.28) is integrated over a control volume dV shown in Fig. 3.2.

$$\int \left[\rho u \frac{\partial \Phi}{\partial x} - \frac{\partial}{\partial x} \left(\Gamma \frac{\partial \Phi}{\partial x}\right)\right] dV = \left[\rho u \Phi_{\rm e} - \left(\Gamma \frac{\partial \Phi}{\partial x}\right)_{\rm e}\right] A_{\rm e} - \left[\rho u \Phi_w - \left(\Gamma \frac{\partial \Phi}{\partial x}\right)_{\rm w}\right] A_{\rm w} \quad (3.29)$$



Figure 3.2 Nomenclature for the discretization in the *x*-direction.

With assumption of the piecewise-linear profile between the grids, the diffusive flux, for example, at the east-face becomes

$$\left(\Gamma \frac{\partial \Phi}{\partial x}\right)_{\rm e} \approx \Gamma_{\rm e} \frac{(\Phi_{\rm E} - \Phi_{\rm P})}{\delta x_{\rm e}} \tag{3.30}$$

with second order accuracy. Other fluxes have a similar form.

Higher-order schemes for discretizing convection terms are used to prevent numerical diffusion which typically occurs with first-order schemes such as the upwind method. First-order schemes lead to pointed bubble shapes in simulations of bubbling fluidized beds (Syamlal, 1998). However, high-order schemes tend to be deficient due to oscillations when discontinuities are encountered. Total variation diminishing (TVD) schemes in the high-order methods can help avoid oscillations. Such schemes use a limiter on the dependent variables (Tannehill et al., 1980). The value of Φ_e at the east face is an example of a TVD scheme and it has the form

$$\Phi_e = \xi_e \Phi_E + \bar{\xi}_e \Phi_P \tag{3.31}$$

where $\bar{\xi}_{e} = 1 - \xi_{e}$.

The convection weighting factor $\xi_{\rm e}$ in Eqn. (3.31) is calculated from the down-wind factor $dw f_{\rm e}$, where

if $u_{\rm e} \ge 0$

$$\Phi_{\rm e} = dw f_{\rm e} \Phi_{\rm E} + (1 - dw f_{\rm e}) \Phi_{\rm P}$$



Figure 3.3 Node locations for the TVD scheme.

 $\xi_{\rm e} = dw f_{\rm e}$

if $u_{\rm e} < 0$

$$\Phi_{\rm e} = dw f_{\rm e} \Phi_{\rm P} + (1 - dw f_{\rm e}) \Phi_{\rm E}$$

$$\xi_{\rm e} = 1 - dw f_{\rm e}$$

The down-wind factor $dw f_{\rm e}$ is calculated using the superbee method which can be applied to explicit conservative convection and diffusion schemes of any order of accuracy (Leonard, 1991)

$$dwf_{\rm e} = \frac{1}{2}\max[0,\min(1,2\theta),\min(2,\theta)]$$

where $\theta = \tilde{\Phi}_{\rm C} / (1 - \tilde{\Phi}_{\rm C})$ and $\tilde{\Phi}_{\rm C}$ is

if $u_{\rm e} \geq 0$

$$\tilde{\Phi}_{\rm C} = \frac{\Phi_{\rm P} - \Phi_{\rm W}}{\Phi_{\rm E} - \Phi_{\rm W}}$$

if $u_{\rm e} < 0$

$$\tilde{\Phi}_{\rm C} = \frac{\Phi_{\rm E} - \Phi_{\rm EE}}{\Phi_{\rm P} - \Phi_{\rm EE}}$$

The locations of $\Phi_{\rm E}$, $\Phi_{\rm W}$, $\Phi_{\rm P}$ and $\Phi_{\rm EE}$ are shown in Fig. 3.3.

The transient term in discretized form is

$$\int \frac{\partial}{\partial t} (\epsilon_m \rho_m \Phi) dV \approx \left[(\epsilon_m \rho_m \Phi)_{\rm P} - (\epsilon_m \rho_m \Phi)_{\rm P}^0 \right] \frac{\delta V}{\delta t}$$
(3.32)

where the superscript 0 indicates previous time step values. Source terms are usually nonlinear and are first linearized as

$$R_{\Phi} \approx \bar{R}_{\Phi} - R'_{\Phi} \Phi_{\rm P} \tag{3.33}$$

where R_{Φ} stands for the constant part of R_{Φ} , while R'_{Φ} is the coefficient of $\Phi_{\rm P}$. Then, the integration of the source term gives

$$\int R_{\Phi} dV \approx \bar{R}_{\Phi} \delta V - R'_{\Phi} \Phi_{\rm P} \delta V$$
(3.34)

For the stability of the iteration, $R'_{\Phi} \geq 0$.

All scalar equations may be arranged to get the following linear equation for Φ

$$a_{\rm P}\Phi_{\rm P} = \sum_{nb} a_{nb}\Phi_{nb} + b \tag{3.35}$$

where b is a source term. Equation (3.35) should be subtracted by the discretized continuity equation, which is multiplied by Φ (Patankar, 1980).

3.2.2.2 Interphase Phase Transfer Terms

The continuity equations for multiphase flows have a source term for interphase mass transfer $\sum R_{lm}$ and can be discretized as

$$\Phi_{\rm P} \sum_{l=0}^{\rm M} R_{lm} = \Phi_{\rm P} \langle R_{lm} \rangle - \Phi_{\rm P} \langle -R_{lm} \rangle$$
(3.36)

where l = 0 indicates gas phase and

$$\langle R \rangle = \begin{cases} 0 & \text{if } R \leq 0 \\ \\ R & \text{if } R > 0 \end{cases}$$

The first term on the right side of Eqn. (3.36) is substituted in the source term b of Eqn. (3.35). $\langle -R_{lm} \rangle$ in the second term of Eqn. (3.36) is substituted in a_P of Eqn. (3.35).

The interphase momentum and energy transfer terms $\sum F_{lm}(u_l - u_m)$ and $\sum \gamma_{lm}(T_l - T_m)$ strongly couple the components of velocity and temperature amongst the phases. These terms should be made fully implicit for solving the equations. However, the equations for each velocity component must be solved together, which leads to a nonstandard matrix structure. The partial



Figure 3.4 Free-slip wall boundary condition.

elimination of interphase coupling (Syamlal, 1998) is used for decoupling the equations.

3.2.2.3 Boundary Conditions

Periodic boundary conditions also referred to as cyclic conditions, are specified for the azimuthal direction in cylindrical coordinates. Rotationally (without pressure drop) or translationally (with pressure drop) cyclic boundary conditions may be specified at any of the boundaries.

The free-slip condition is shown schematically in Fig. 3.4 for a cell abut a wall and the ghost cell. The free-slip requirement is

$$(V)_{\text{ghost cell}} = (V)_{\text{near wall cell}}$$

so that the gradient of the velocity at the wall is zero. The boundary condition becomes

$$V(i) - V(i-1) = 0 \tag{3.37}$$



Figure 3.5 No-slip wall boundary condition.

The no-slip condition shown schematically in Fig. 3.5 is set as

 $(V)_{\text{ghost cell}} = -(V)_{\text{near wall cell}}$

so that the velocity at the wall is zero. The boundary condition becomes

$$V(i) + V(i-1) = 0 (3.38)$$

3.2.3 Modified SIMPLE Method

The semi-implicit method for pressure-linked equations (SIMPLE) was developed for the calculation of the single flow field and it has served well (Patankar, 1980). Here, an extension of SIMPLE is used to solve the discretized equations. However, the method requires modifications for multiphase flows due to more field variables and more equations.

The SIMPLE algorithm derives the pressure correction equation to handle the pressure terms in the momentum equations. These pressure terms are not expressed explicitly using an equation. Velocities solved from the momentum equations with the correct pressure satisfy the continuity equations. Similar procedures can be applied to multiphase flows using solid pressure corrections. However, the solid volume fraction correction equation is used instead of solid pressure correction equation since solid pressure correction equation requires that $\partial P_s / \partial \epsilon_s$ does not vanish when $\epsilon_s \to 0$.

In densely packed regions, the solid pressure is an exponentially increasing function of the solid volume fraction. A small increase in the solid volume fraction will cause a large increase in the solid pressure. For numerical stability, solid volume fraction corrections are underrelaxed, where

If
$$(\epsilon_m)_{\text{new}} > (\epsilon)_p$$
 and $\epsilon'_m > 0$
 $\epsilon'_m = \omega_{\epsilon_m} \epsilon'_m$
(3.39)

and ϵ_p is the volume fraction for the packed region, ω_{ϵ_m} is an under relaxation factor, and ϵ'_m is correction of solid volume fraction with guessed value ϵ^*_m

$$\epsilon_m = \epsilon_m^* + \epsilon_m' \tag{3.40}$$

A transient simulation is performed since gas solid flows are inherently unstable. Small time steps for a simulation and underrelaxation for iteration are chosen to ensure the numerical stability. Further numerical details and parameters will be discussed with results.

CHAPTER 4. IMPLEMENTATION OF CHEMICAL REACTIONS USING IN-SITU ADAPTIVE TABULATION

A wide range of time scales involved in typical kinetic schemes requires specialized treatment of chemical source terms. Available approaches include reduction and storage and retrieval (§2.2). In-situ adaptive tabulation (ISAT) is a storage and retrieval strategy, which has been efficiently used to treat complex chemistry (Saxena and Pope, 1999; James et al., 1999) and is readily adapted to any time-marching CFD code using fractional time stepping. In this chapter, the ISAT algorithm and implementation of ISAT into MFIX are presented in detail.

4.1 In-Situ Adaptive Tabulation Applied to Chemical Reaction Calculation

4.1.1 Decoupling Chemical Source Terms from Transport Terms

In a gas-solid mixture, the thermochemical state can be determined by $N_{\rm s}$ variables such as mass fractions and enthalpy. However, if there are other dependencies existing between these variables, the degrees of freedom decrease to D. The composition Φ is defined to include Dvariables, which is a subset of total $N_{\rm s}$ variables. The transport equation for the composition has the form

$$\frac{d\Phi}{dt} = S(\Phi) + T(\Phi)$$
(4.1)

where Φ has components $\phi_1, \phi_2, \dots, \phi_D$, $S(\Phi)$ and $T(\Phi)$ are the changes in Φ due to chemical reactions and transport, respectively. The composition of reactive flows is affected by reactions and transport which includes convection and diffusion as shown in Eqn. (4.1). Therefore, over a small time step δt , the change of composition when using fractional time stepping becomes

$$\Phi(t) \stackrel{\text{transport,}}{\longrightarrow} \Phi(t + \delta t)$$

where $\Phi(t)$ and $\Phi(t + \delta t)$ are the compositions at time t and $t + \delta t$, respectively.

When CFD codes use fractional time stepping (e.g., MFIX) for reactive flow calculations, splitting techniques can be applied so that the different processes can be treated in separate fractional steps. In the first fractional time step, the change due to convection and diffusion is solved for every node using equations

$$\frac{d\Phi}{dt} = T(\Phi) \tag{4.2}$$

with the initial condition $\Phi(t)$. The solution of Eqn. (4.2) is denoted as $\Phi'(t + \delta t)$. In the next fractional time step, the change due to chemical reactions is solved by

$$\frac{d\Phi}{dt} = S(\Phi) \tag{4.3}$$

with the initial condition $\Phi'(t + \delta t)$. The composition of all nodes are then approximated to $\Phi(t + \delta t)$. Usually, the change due to the reactions is calculated last since that some of the reaction time scales are smaller than the flow time scales (Fox, 2003).

The overall fractional time stepping is represented as

$$\Phi(t) \xrightarrow{\text{transport}} \Phi'(t+\delta t) \xrightarrow{\text{reactions}} \Phi(t+\delta t)$$

Therefore, the chemical source term is decoupled from the transport term and can be treated with efficient numerical methods such as ISAT. Thus the problem statement for solving chemical source terms is based on a given the chemical composition at time t to determine the composition at time $t + \delta t$ resulting from chemical reactions. Equation (4.3) is integrated repetitively for each node at every time step with each new set of initial conditions.

4.1.2 Overview of ISAT Algorithm

ISAT has some characteristics to overcome the disadvantages of pre-computed look-up tables (Fox, 2003). For example, the method can efficiently calculate detailed chemistry. The in-situ nature reduces the tabulation to only the accessed region which is much smaller than the realizable region in which reactions can occur. An adaptive algorithm is employed to control interpolation errors while minimizing the number of points tabulated. Furthermore, the binary-tree tabulation algorithm is used to generate tables and access data. These characteristics are introduced in detail as follows.

4.1.2.1 Linearized Mapping

A non-linear reaction map is defined by integrating Eqn. (4.3) over an arbitrary time δt with an initial condition Φ_0

$$\Phi(\delta t) = R(\Phi_0; \delta t) \tag{4.4}$$

Thus the reaction mapping is a system of ordinary differential equations (ODEs)

$$\frac{\partial R(\Phi_0; \delta t)}{\partial t} = S(R(\Phi_0; \delta t)) \tag{4.5}$$

with $R(\Phi_0; 0) = \Phi_0$. The mapping Jacobian matrix, which represents the sensitivity of the reaction mapping to changes in the initial conditions, is defined as

$$A(\Phi_0; \delta t) = \frac{\partial R(\Phi_0; \delta t)}{\partial \Phi_0}$$
(4.6)

Differentiating Eqn. (4.5) with respect to Φ_0

$$\frac{\partial^2 R(\Phi_0; \delta t)}{\partial t \partial \Phi_0} = \frac{\partial S(R(\Phi_0; \delta t))}{\partial \Phi_0} \tag{4.7}$$

Substituting Eqn. (4.6) into Eqn. (4.7) and using the chain rule

$$\frac{\partial A(\Phi_0;\delta t)}{\partial t} = \frac{\partial S(R(\Phi_0;\delta t))}{\partial R(\Phi_0;\delta t)} \frac{\partial R(\Phi_0;\delta t)}{\partial \Phi_0}$$
(4.8)

The matrix A of Eqn. (4.8) can be expressed by

$$\frac{\partial A(\Phi_0;\delta t)}{\partial t} = J(R(\Phi_0;\delta t))A(\Phi_0;\delta t)$$
(4.9)

with $A(\Phi_0; 0) = I$, where I is the identity matrix. The chemical-source-term Jacobian matrix J is defined as

$$J(\Phi) = \frac{\partial S(\Phi)}{\partial \Phi}$$

Therefore, the values of $\Phi(\delta t)$ and $A(\Phi_0; \delta t)$ can be calculated by integrating Eqs. (4.5) and (4.9) with an initial composition Φ_0 and the time step δt . This step is referred to as direct integration (DI).

The ISAT contains information, Φ_0 , $\Phi(\delta t)$ and $A(\Phi_0; \delta t)$, for each tabulation point. For a query point Φ_0^q , $\Phi^q(\delta t)$ is calculated by linear interpolation from a neighboring point $\Phi(\delta t)$

$$\Phi^{q}(\delta t) = \Phi(\delta t) + A(\Phi_{0}; \delta t)(\Phi_{0}^{q} - \Phi_{0})$$
(4.10)

instead of by direct integration, where

$$\Phi_{\rm DI}^{\rm q}(\delta t) = R(\Phi_0^{\rm q}; \delta t) \tag{4.11}$$

Thus the error induced by the linear approximation is

$$\epsilon_{\rm L} = \Phi_{\rm DI}^{\rm q}(\delta t) - \Phi^{\rm q}(\delta t)$$

The linearized mapping for a query point Φ_0^q is shown in Fig. 4.1 with $\delta \Phi^q = \Phi^q(\delta t) - \Phi(\delta t)$ and $\delta \Phi_0^q = \Phi_0^q - \Phi_0$.

4.1.2.2 Ellipsoid of Accuracy and Binary Tree

The ellipsoid of accuracy (EOA) is used to control the interpolation errors, which are stored in a specified number of binary trees. Each leaf of a tree is an EOA. Each call for ISAT is



Figure 4.1 The linearized mapping used in ISAT.

referred to as a query. Thus for a given query point Φ_0^q , one of the following will occur:

- If an EOA containing the query point is found, the linear approximation is returned. This is called *retrieve*.
- 2. If the query is not fulfilled by a retrieve, then a direct integration is performed. Based on the value of direct integration, the EOA is grown to include the query point if the linear approximation is sufficiently accurate. This outcome is referred to as a *grow*.
- If the query is not satisfied by a grow, a new EOA is added. The outcome is referred to as an *add*.
- 4. If the table is full, an old EOA is deleted, and the new one is added; or no new EOA's are added, and the query is resolved by DI. This is called *replace*.

4.1.2.3 Flow Chart of ISAT Algorithm

Figure 4.2 shows the key steps in the ISAT algorithm. At the beginning of the simulation, the binary tree is empty and thus the first call begins at the point marked "FIRST QUERY". Then, all calls to the algorithm begin at the point marked "NEXT QUERY". The next step is to check the EOA, as described in the previous section. If the query point is outside of



Figure 4.2 The flow chart of the key steps in ISAT.

the EOA, direct integration is performed and then either the EOA grows or a new EOA is created. Otherwise, if the query point is inside the EOA, linear approximation is returned and the table is updated. For best performance, nearly all calls result in a "Retrieve", thereby avoiding direct integration.

4.2 Implementation of ISAT in MFIX

4.2.1 Decoupling Chemical Source Terms from Transport Terms in MFIX

The continuity, species and energy equations formulated in MFIX were presented in §3.1.1. Recall that the change of volumetric fractions, gas and solid temperatures and mass fractions are affected by transport and reactions. For every fractional time step, the solution flow chart



Figure 4.3 The flow chart of original MFIX.

for original MFIX is shown in Fig. 4.3. During time marching, the velocities and volumetric fractions are calculated first. Then the temperatures and species mass fractions are solved. At last, the residuals are checked for convergence. If the convergence criterion is satisfied, the simulation goes to the next time step. Otherwise, the iterations are continued.

Thus, the equations are integrated over a computational cell in the finite-volume method. Usually, the chemistry is stiff, for example, due to the large range of time scales. The equations are tightly coupled during the iterations of each time step. Therefore, a time-splitting method can be employed to solve the equations.

Applying time-splitting techniques to MFIX, the calculation of species mass fractions, temperatures and volumetric fractions requires two steps. 1. In the first fractional time step, the change due to convection is solved for every node using the Eqs. (3.1), (3.2) and (3.5)–(3.8) by setting the chemical source terms to zero

$$\frac{\partial}{\partial t}(\epsilon_{\rm g}\rho_{\rm g}) + \nabla \cdot (\epsilon_{\rm g}\rho_{\rm g}\vec{v}_{\rm g}) = 0, \qquad (4.12)$$

$$\frac{\partial}{\partial t}(\epsilon_{\rm sm}\rho_{\rm sm}) + \nabla \cdot (\epsilon_{\rm sm}\rho_{\rm sm}\vec{v}_{\rm sm}) = 0, \qquad (4.13)$$

$$\epsilon_{\rm g}\rho_{\rm g}C_{\rm pg}\left(\frac{\partial T_{\rm g}}{\partial t} + \vec{v}_{\rm g}\cdot\nabla T_{\rm g}\right) = -\nabla\cdot\vec{q}_{\rm g} - \sum_{m=1}^{M}H_{\rm gsm} + H_{\rm wall}(T_{\rm wall} - T_{\rm g}),\tag{4.14}$$

$$\epsilon_{\rm sm}\rho_{\rm sm}C_{\rm psm}\left(\frac{\partial T_{\rm sm}}{\partial t} + \vec{v}_{\rm sm} \cdot \nabla T_{\rm sm}\right) = -\nabla \cdot \vec{q}_{\rm sm} + H_{\rm gsm},\tag{4.15}$$

$$\frac{\partial}{\partial t} (\epsilon_{\rm g} \rho_{\rm g} X_{\rm gn}) + \nabla \cdot (\epsilon_{\rm g} \rho_{\rm g} X_{\rm gn} \vec{v}_{\rm g}) = \nabla \cdot D_{\rm gn} \nabla X_{\rm gn}, \qquad (4.16)$$

$$\frac{\partial}{\partial t} (\epsilon_{\rm sm} \rho_{\rm sm} X_{\rm smn}) + \nabla \cdot (\epsilon_{\rm sm} \rho_s X_{\rm smn} \vec{v}_{\rm sm}) = \nabla \cdot D_{\rm smn} \nabla X_{\rm smn}, \qquad (4.17)$$

with the initial conditions $X_{gn}(t)$, $X_{smn}(t)$, $\epsilon_{sm}(t)$, $\rho_g(t)$, $T_g(t)$ and $T_{sm}(t)$. The solutions of Eqs. (4.12)–(4.17) are $X'_{gn}(t + \delta t)$, $X'_{smn}(t + \delta t)$, $\epsilon'_{sm}(t + \delta t)$, $\rho'_g(t + \delta t)$, $T'_g(t + \delta t)$ and $T'_{sm}(t + \delta t)$. For clarity, the spatial dependency has been suppressed. However, the reader should keep in mind that the variables must be updated in time at every grid cell.

2. In the next fractional time step, the change due to the chemical source terms for mass fractions are

$$\frac{\mathrm{d}}{\mathrm{d}t}(\epsilon_{\mathrm{g}}\rho_{\mathrm{g}}X_{\mathrm{g}n}) = R_{\mathrm{g}n} \tag{4.18}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}(\epsilon_{\mathrm{sm}}\rho_{\mathrm{sm}}X_{\mathrm{smn}}) = R_{\mathrm{smn}} \tag{4.19}$$

Differentiating Eqs. (4.18) and (4.19) and rearranging terms yields

$$\frac{\mathrm{d}}{\mathrm{d}t}(X_{\mathrm{g}n}) = \frac{R_{\mathrm{g}n}}{\epsilon_{\mathrm{g}}\rho_{\mathrm{g}}} - \frac{X_{\mathrm{g}n}}{\epsilon_{\mathrm{g}}\rho_{\mathrm{g}}} \frac{\mathrm{d}}{\mathrm{d}t}(\epsilon_{\mathrm{g}}\rho_{\mathrm{g}}), \qquad (4.20)$$

$$\frac{\mathrm{d}}{\mathrm{d}t}(X_{\mathrm{smn}}) = \frac{R_{\mathrm{smn}}}{\epsilon_{\mathrm{sm}}\rho_{\mathrm{sm}}} - \frac{X_{\mathrm{smn}}}{\epsilon_{\mathrm{sm}}\rho_{\mathrm{sm}}} \frac{\mathrm{d}}{\mathrm{d}t}(\epsilon_{\mathrm{sm}}\rho_{\mathrm{sm}}), \qquad (4.21)$$

where the change of volumetric fractions ϵ_g and ϵ_{sm} are calculated from the continuity equations for the solid phases

$$\frac{\mathrm{d}}{\mathrm{d}t}(\epsilon_{\mathrm{sm}}\rho_{\mathrm{sm}}) = \sum_{n=1}^{N_{\mathrm{sm}}} R_{\mathrm{smn}} \tag{4.22}$$

with $\epsilon_{\rm g} + \sum_{m=1}^{M} \epsilon_{{\rm s}m} = 1$. Eqn. (4.22) can be rearranged as

$$\frac{\mathrm{d}}{\mathrm{d}t}(\epsilon_{\mathrm{s}m}) = \frac{1}{\rho_{\mathrm{s}m}} \sum_{n=1}^{N_{\mathrm{s}m}} R_{\mathrm{s}mn} \tag{4.23}$$

since $\rho_{\mathrm sm}$ is constant. Beginning with the continuity equation for the gas phase

$$\frac{\mathrm{d}}{\mathrm{d}t}(\epsilon_{\mathrm{g}}\rho_{\mathrm{g}}) = \sum_{n=1}^{N_{\mathrm{g}}} R_{\mathrm{g}n}$$
(4.24)

the equation can be rewritten

$$\epsilon_{\rm g} \frac{\mathrm{d}\rho_{\rm g}}{\mathrm{d}t} - \rho_{\rm g} \frac{\mathrm{d}\epsilon_{\rm s}}{\mathrm{d}t} = \sum_{n=1}^{N_{\rm g}} R_{\rm gn} \tag{4.25}$$

Then, the change due to gas density is

$$\frac{\mathrm{d}}{\mathrm{d}t}(\rho_{\mathrm{g}}) = \frac{1}{(1-\epsilon_{\mathrm{s}})} \left(\sum_{n=1}^{N_{\mathrm{g}}} R_{\mathrm{g}n} + \rho_{\mathrm{g}} \frac{\mathrm{d}\epsilon_{\mathrm{s}}}{\mathrm{d}t} \right)$$
(4.26)

The changes due to the chemical reactions for temperatures are

$$\epsilon_{\rm g} \rho_{\rm g} C_{\rm pg} \frac{\mathrm{d}T_{\rm g}}{\mathrm{d}t} = -\Delta H_{\rm rg} \tag{4.27}$$

$$\epsilon_{\rm sm}\rho_{\rm sm}C_{\rm psm}\frac{\mathrm{d}T_{\rm sm}}{\mathrm{d}t} = -\Delta H_{\rm rsm} \tag{4.28}$$

The equations can be rearranged as

$$\frac{\mathrm{d}T_{\mathrm{g}}}{\mathrm{d}t} = \frac{-\bigtriangleup H_{\mathrm{rg}}}{\epsilon_{\mathrm{g}}\rho_{\mathrm{g}}C_{\mathrm{pg}}} \tag{4.29}$$

$$\frac{\mathrm{d}T_{\mathrm{s}m}}{\mathrm{d}t} = \frac{-\bigtriangleup H_{\mathrm{rs}m}}{\epsilon_{\mathrm{s}m}\rho_{\mathrm{s}m}C_{\mathrm{ps}m}} \tag{4.30}$$

Thus Eqs. (4.20) and (4.21) for mass fractions, Eqn. (4.23) for volumetric fractions, Eqn. (4.26) for gas density and Eqs. (4.29) and (4.30) for temperatures are integrated with the initial condition $X'_{gn}(t + \delta t)$, $X'_{smn}(t + \delta t)$, $\epsilon'_{sm}(t + \delta t)$, $\rho'_{g}(t + \delta t)$, $T'_{g}(t + \delta t)$ and $T'_{sm}(t + \delta t)$. The mass fractions, volumetric fractions, gas density and temperatures at all nodes are updated to $X_{gn}(t + \delta t)$, $X_{smn}(t + \delta t)$, $\epsilon_{sm}(t + \delta t)$, $\rho_{g}(t + \delta t)$, $T_{g}(t + \delta t)$, $T_{sm}(t + \delta t)$ and pressure is updated using the ideal gas law . In order to use an ODE solver, the user must provide the RHS of Eqs. (4.20), (4.21), (4.23), (4.26), (4.29) and (4.30) and their Jacobian matrix.

With the time-splitting method the chemical reactions for the gas and solid phases are isolated from the transport terms and can be treated with efficient numerical methods such as ISAT. The problem statement for solving the chemical source terms is based on a given chemical composition at time t to determine the composition at time $t + \delta t$ resulting from chemical reactions. During the course of a simulation, Eqs. (4.20), (4.21), (4.23), (4.26), (4.29) and (4.30) are integrated repetitively for each node at every time step with each new set of initial conditions.

Thus, as shown in Fig. 4.4, the chemical source terms are solved using ISAT and the transport terms are calculated using MFIX at each fractional time step. The change in species mass fractions, volumetric fractions and temperatures due to transport is calculated with other equations using iterations in the MFIX code. If the equations converge, the change in species mass fractions, volumetric fractions and temperatures due to chemical reactions are computed using ISAT. Then the calculation moves to the next time step. In order to ensure rapid convergence, MFIX adapts the time step $\delta t_{\rm M}$ on every iteration.



Figure 4.4 The flow chart of MFIX with ISAT.

4.2.2 ISAT User Subroutines

4.2.2.1 Direct Integration and Mapping Jacobian Matrix

As discussed in §4.1.2, direct integration of Eqs. (4.20), (4.21), (4.23), (4.26), (4.29) and (4.30) should be provided to ISAT. In this research, Rosenbrock (Press et al., 1992) and ODEPACK (Stepleman, 1983) stiff ODE solvers are used for integration. The chemical-source-term Jacobian matrix is calculated numerically using ADIFOR 2.0 (Bischof et al., 1998).

The mapping Jacobian matrix A can be calculated by integrating Eqn. (4.9) with the initial condition A(0) = I.

$$A(\delta t) = A(0)e^{J\delta t} \tag{4.31}$$

A subroutine computes the matrix exponential of a general full matrix is available in Numerical Recipes (Press et al., 1992). The subroutine uses an irreducible rational Pade approximation for computing A.

4.2.2.2 ISAT Control Parameters

The selection of control parameters are crucial for effective ISAT usage. The following is a description of the control parameters.

- Scaling: The positive scale factors can be assigned to variables, which can improve ISAT performance. However, the elements of the scaled mapping gradient matrix A should not be larger than unity.
- 2. Transformations of variables: Due to the use of linear approximation, ISAT is most effective if the chemical-source-term is a linear function of Φ . Thus, a transformation of variables can be considered to reduce the level of non-linearity.
- 3. Error tolerance: The specification of the error tolerance is crucial for ISAT which is used to control the interpolation error. Therefore, a smaller value of error tolerance will yield better results. However, with lower error tolerance, a larger table is generated and the number of retrieves decreases, thus time for calculation increases. A larger value of error tolerance generates less accurate results with lower CPU time.
- 4. Other parameters are specified in ISAT manual such as the number of binary trees used in the tabulation and the maximum allowable table size. Normally, the maximum allowable table size should be set large enough that the limit is never reached.

CHAPTER 5. SIMULATIONS OF FLUIDIZED BED PYROLYSIS OF SILANE

In-situ adaptive tabulation (ISAT) has some advantages over the other storage and retrieval methods. It is readily adapted to any time-marching CFD code using fractional time stepping, e.g. MFIX. The implementation of ISAT into MFIX is presented in §4.2 in detail. In this chapter, the accuracy and performance of ISAT with MFIX are investigated with a silane pyrolysis case. The numerical procedure is presented with isothermal simulations. The influence of scale variables and multiple tables is introduced using non-isothermal simulations.

5.1 Introduction

Silane pyrolysis is an example of chemical vapor deposition (CVD). CVD is a method to modify the surface state of the particles which influences particle properties such as corrosion resistance and flowability. Each particle is coated by a thin layer of new material, thereby changing the surface condition. A fluidized bed is used for CVD due to its isothermal and thoroughly mixed particle properties resulting in uniformity of surface change. Two applications of silane pyrolysis in industry are: protecting either the particles or surfaces immersed in a fluidized bed from oxidation or corrosion by depositing silicon films on them, and, producing ultrapure silicon for microelectronic and photovoltaic uses by growing the fluidized silicon particles.

Detailed chemical reactions for simulations of silane pyrolysis are considered (Caussat et al., 1995a,b). A fluidized bed reactor is fed with a mixture of silane (SiH₄) and nitrogen (N₂). First, a reversible, gas-phase reaction occurs:

$$SiH_4 \leftrightarrow SiH_2 + H_2 + 238.5 \text{ kJ/mol}$$

where highly reactive SiH_2 undergoes a gas-phase reaction to form Si_2H_6 :

$$SiH_2 + SiH_4 \iff Si_2H_6 - 227.2 \text{ kJ/mol}$$

Then the heterogeneous decomposition of SiH_4 and SiH_2 on alumina (Al₂O₃) particles is described by two irreversible reactions:

$$\begin{split} \mathrm{SiH}_4 & \longrightarrow \mathrm{Si(s)} + 2\mathrm{H}_2 - 34.3 \ \mathrm{kJ/mol} \\ \mathrm{SiH}_2 & \longrightarrow \mathrm{Si(s)} + \mathrm{H}_2 - 272.8 \ \mathrm{kJ/mol} \end{split}$$

The overall reaction involves five gaseous species (SiH₄, H₂, Si₂H₆, SiH₂ and N₂) and two solid species (Al₂O₃ and Si) which need to be modeled in a numerical simulation. To determine the production and consumption of the gases, the kinetic constants of Faylle (1993) have been selected. The heterogeneous reactions of silane on surfaces follow the kinetic laws of Furusawa et al. (1988) and Guenther et al. (2001). The thermodynamic data of silicon hydrides are obtained from Gernot et al. (1997).

5.2 Isothermal Simulations

Figure 5.1 shows the two-dimensional (2D) geometry of a silane pyrolysis reactor, and two different domain sizes for this study are given in Tab. 5.1. An experimental and theoretical study for silane pyrolysis in a fluidized bed (Case 1) was performed by Caussat et al. (1995a,b). Guenther et al. (2001) numerically simulated the experiment at a fixed temperature using MFIX without time-splitting, and the results showed excellent agreement with experiments.

In simulations, the solid particles in the bed are Al_2O_3 with density 3.9 g/cm³ and diameter 8.2×10^{-3} cm. A mixture of SiH₄ and N₂ enters from the bottom of the bed with a velocity of 7.89 cm/s. The outlet boundary condition is set to a constant pressure of 1.0×10^5 Pa.



Figure 5.1 The computational 2D domain for fluidized bed pyrolysis of silane.

Table 5.1 The domain sizes (cm) for silane pyrolysis reactors.

Case	R1	R2	R3	H1	H2	H3	H4
1	5	2.65	2	33.2	63	74	80
2	5	2.65	2	12.0	13	24	30

The temperature inside the bed and at the boundaries is 882.45 K. At the center line, freeslip boundary conditions are set for both the gas and solid phases, while no-slip boundary conditions are set at the wall. The minimum void fraction in the bed is 0.5246 and a cylindrical coordinate system is used for the simulations. The number of cells is 25 and 80 in the radial and axial directions, respectively, which is determined to be grid independent by Guenther et al. (2001). For the isothermal case with chemical reactions, the continuity, species and momentum equations for the gas and solid phases are solved without the energy equation. All simulations were performed on Sunfire 6800 using a single processor with ISAT (version 3.0). For DI of equations, the source terms and Jacobian matrix of the source terms are provided to a Rosenbrock stiff ODE solver (Press et al., 1992).

5.2.1 Numerical Procedure for MFIX With ISAT

As mentioned in §4.2, ISAT is called by MFIX at every time step with a time increment, denoted as $\delta t_{\rm M}$. The mass fractions, volumetric fractions and temperatures which are transfered from MFIX to ISAT, are updated after the time step $\delta t_{\rm M}$ by DI or linear interpolation, and the values are returned to MFIX to start the next iteration.

The results denoted by ISAT(DI) are found using time-splitting where the calls to ISAT use DI only. No linear interpolation is used for the ISAT(DI) calculations. These results are compared with the results of MFIX to test the accuracy of the time-splitting method. In the tests using linear interpolation, an error tolerance (E) of 10^{-5} is chosen to show the full capabilities of ISAT. This error tolerance was obtained by testing silane decomposition simulations using the global chemical mechanism (Furusawa et al., 1988; Xie, 2002). The domain size of Case 1 was used for the first set of simulations with ISAT. ISAT began with an empty table and all scale factors were chosen as one.

Figure 5.2 shows plots of radial void fraction and mass fraction of SiH₄ averaged over 10-75 seconds for MFIX, ISAT(DI), and ISAT($E=10^{-5}$). The data are averaged only over a height of 2-10 cm since the chemical reactions occur mainly in the lower region of the bed. The bubbles, which are indicated by a high void fraction (Fig. 5.2(a)), are away from the centerline due to the fact that the simulations are two-dimensional (Guenther et al., 2001). The high mass fraction of SiH₄ (Fig. 5.2(b)) appears near the wall (r=2 cm). In general, ISAT(DI) is in very good agreement with MFIX as shown in Fig. 5.2. Therefore, the time-splitting method of MFIX with ISAT is successfully implemented. Based on Fig. 5.2, the averaged data for ISAT(DI) and ISAT($E=10^{-5}$) also compare well confirming that the error tolerance is accurate enough for the simulation of silane pyrolysis.

ISAT performance for the error tolerance of 10^{-5} is summarized in Tab. 5.2, listing the percentage of CPU time used by *retrieves*, grows and adds. Although the percentage of CPU time for *retrieves* is large, it is still too small for good performance of ISAT, which needs almost all query points to be *retrieves*. In addition, the number of records is relatively large compared to the number of queries. Large records means that *retrieves* do not dominate the



Figure 5.2 (a) Radial void fraction and (b) mass fraction of SiH₄ averaged over 10–75 s and height 2-10 cm using MFIX, ISAT(DI) and ISAT($E=10^{-5}$).



Figure 5.3 CPU time for ISAT events versus queries.

Table 5.2 A summary of ISAT performance for silane decomposition with an error tolerance of $E=10^{-5}$ for Case 1.

Percentage of time	%
Retrieves	95
Grows	4
Adds	1
Number of queries	$5.4 imes 10^7$
Records	$6.2 imes 10^5$

ISAT calculation. A graph of ISAT performance is provided in Fig. 5.3 and reflects the poor performance of ISAT as shown in Tab. 5.2. Note that the *add* and *grow* curves in Fig. 5.3 do not flatten for large numbers of queries. Therefore, the table is still growing and records are being added to the table, which indicates that the table has not reached maturity at the end of the simulation. This behavior was unexpected based on the results of other applications of ISAT for reactive flows. We were thus motivated to determine its cause.

As noted earlier, MFIX calls ISAT at each fractional time step $\delta t_{\rm M}$, which changes erratically during the course of the simulation. A plot of the time step $\delta t_{\rm M}$ versus time is shown in Fig. 5.4 for the silane pyrolysis using MFIX. The time step increases at the beginning of the



Figure 5.4 Time step versus time for silane decomposition.

simulation, but becomes statistically steady after about 10 seconds. However, the time step fluctuates markedly around the mean value, and is essentially uncorrelated in time.

The poor performance of the ISAT is due to this drastically varying time step in MFIX. Since ISATAB determines the EOA based both on the distance in the composition space and the difference in the time step, a variable time step in ISAT with similar mass fractions and volumetric fractions will produce a new record or growth instead of utilizing the linear interpolation procedure. It appears that a CFD code with a rapidly varying time step needs longer time and more memory to build and store the ISAT table. One possible remedy is to call ISAT from MFIX at a fixed time step $\delta t_{\rm I}$ during the calculation as shown in Fig. 5.5. At the beginning, $t_{\rm M}$ is equal to $t_{\rm I}$. Time steps in MFIX ($\delta t_{\rm M}$) vary during the simulation and ISAT is only called when the time in MFIX advances more than $\delta t_{\rm I}$. Then, ISAT updates the data every $\delta t_{\rm I}$, which is held constant until $t_{\rm I}$ is greater than $t_{\rm M}$.

5.2.2 Results of 2D Simulations Using a Fixed Time Step

With the same numerical procedure discussed in §5.2.1, silane pyrolysis was again used to test the accuracy and speed-up of ISAT with different number of trees. In simulations, $t_{\rm I}$ is set as 10^{-3} , which is a value smaller than the averaged time step obtained by DI shown in Fig 5.4.



Figure 5.5 Flow chart of MFIX with ISAT for fixed time step.

However, the height of the computational domain was reduced (Case 2 in Tab. 5.1) to 30 cm in order to decrease the overall computational time and increase the percentage of CPU time used for the chemical reaction calculation.

All ISAT calculations used the same error tolerance $(E=10^{-5})$ as before. Different numbers of trees, 2 and 10, were used to test the performance of ISAT, and results for ISAT(DI) and ISAT with a fixed time step are shown in Fig. 5.6. Good agreement is observed for void fraction and mass fraction of SiH₄ between ISAT(DI) and ISAT, and the number of trees has a negligible effect on the accuracy.

ISAT performance is summarized in Tab. 5.3. Total CPU time is the time used for the entire MFIX–ISAT calculation, whereas the CPU time for chemistry is the time used only by ISAT. The speed-up of chemistry is the ratio of CPU time between ISAT(DI) and ISAT for the chemistry calculation. For the sake of comparison, also included in the table are results using ISAT ($E=10^{-5}$) allowing the calculations to proceed with a variable time step. Notably, the percent retrieves is poor due to the enormous number of records. Thus, although the number of



Figure 5.6 (a) Radial void fraction and (b) mass fraction of SiH₄ averaged over 100–200 s and height 2-10 cm using ISAT(DI), ISAT(ntree=2) and ISAT(ntree=10) with $E=10^{-5}$.



Figure 5.7 CPU time for ISAT events versus queries for ntree=10.

queries with a variable time step is 34% less, the CPU time for chemistry increases by a factor of 5.8. ISAT using a fixed time step reduces the computational time for chemistry drastically and the speed-up is around 46 for silane pyrolysis. Moreover, the computational time decreases when the number of trees increases. However, the total CPU time does not reduce much due to the small percentage of computational time occupied by the chemistry calculation. Comparing the results for ntree equal to 2 and 10, as the number of trees increases, CPU time for the chemistry calculation decreases since the depth of tree is less and the time for a

Table 5.3 A summary of ISAT performance for isothermal simulations.

	$E = 10^{-5}$			
	ntree=10	ntree = 10	ntree=2	
	(variable $\delta t_{\rm I}$)	(fixed $\delta t_{\rm I}$)	(fixed $\delta t_{\rm I}$)	DI
Total CPU time (s)	187771	160927	169394	202533
CPU time for chemistry (s)	6269	1091	1353	50723
Number of queries	2.88×10^7	4.37×10^7	4.37×10^7	
Retrieves $(\%)$	95.12	99.93	99.49	
Records	$2.4 imes 10^5$	569	5102	
Speed-up of chemistry	5.5	46.5	37.5	

retrieve is shorter. Figure 5.7 is a graphical representation of Tab. 5.3 for ntree=10. As seen in Fig. 5.7, a newly created table is dominated by *adds*, followed by an intermediate period dominated by *grows*. When *adds* and *grows* become flat, the overall computational efficiency of ISAT increases dramatically since the table is mature and almost every query point leads to a *retrieve*. Finally, note that with a fixed time step the number of records is drastically reduced as compared with a variable time step. This is an indication that ISATAB does not perform efficiently with a rapidly varying time step.

5.3 Non-Isothermal Simulations

Version 4.0 of ISAT was released in 2003 with the capability to be implemented into parallel codes using MPI. A parallel code for multiprocessors can reduce the computational time drastically, which is important for multiphase flow calculations since the simulations are computationally expensive due to small time steps and grid size chosen to capture flow scales. The addition of chemistry and interphase mass transport into the calculations adds to the computational expense too. Another feature of ISAT 4.0 is that it can be used to construct distinct tables for different functions on each processor. Also, secondary retrieves and secondary grows are added into the algorithm. In the primary retrieve, the trees are traversed in turn to reach the primary-retrieve leaf. If the value can be retrieved from this leaf, then this value is immediately returned. If the primary retrieve fails, the trees are traversed again and a secondary retrieve from each tree is attempted. If the value can be retrieved from any leaf tested, then this value is returned. If the secondary retrieve fails, then the value is evaluated by DI, and it is attempted to grow the primary-retrieve leaf of each tree. If primary grow fails, the trees are traversed in turn and a number of leaves are grown if possible by secondary grows.

With the new capabilities of ISAT, non-isothermal silane cases were simulated to test the performance of ISAT. The reaction kinetics and properties of gas and solid phases are shown in §5.2. However, energy equations for gas and solids were solved along with the continuity, momentum and species equations. The initial condition for the temperatures of gas and solids in the bed and gas at the inlet is 882.45 K. Fixed temperature conditions (882.45 K) are set at

Case	Variables	Scale factors	ntree	Е
1	All	1.0	16	10^{-5}
	$T_{ m g}$	882 K		
	$T_{\mathrm sm}$	882 K		
	$X_{ m SiH_4}$	0.15		
	$X_{\rm SiH_2}$	10^{-6}		
2	X_{H_2}	0.05	16	10^{-2}
	$X_{\rm Si2H_6}$	0.05		
	$X_{\rm Si}$	5×10^{-3}		
	$ ho_{ m g}$	10^{-4}		
	All others	1.0		

Table 5.4 ISAT control parameters for 2D non-isothermal simulations.

walls for both phases. All calculations were performed on an Alpha Cluster (HPC3) at Iowa State University. The direct integration of ODEs is calculated by ODEPACK (Stepleman, 1983).

5.3.1 2D Simulations Using Scale Factors

As mentioned in §4.2.2.2, positive scale factors can be assigned to the independent and the dependent variables. The guideline is that the elements of the scaled gradient matrix should not be large compared to unity. Thus ISAT is tabulated in terms of the normalized quantities such that the non-isothermal 2D cases are simulated with scale factors. The computational domain in the simulations is the same as the isothermal 2D cases shown in Fig. 5.1.

The control parameters for two different sets of scale factors are listed in Tab. 5.4. The same number of trees (16) were used in both cases. Case 1 sets all scale factors as the default values which are unity, which implies that variables are not scaled. Case 2 scales the temperatures by the inlet gas and wall temperature which is 882 K. All other scale factors are the maximum values appearing in the simulations obtained by DI. The factors are shown in Tab. 5.4. Numerical tests showed that an error tolerance of 10^{-5} for case 1 yields satisfactory accuracy with DI. However, case 2 can use a larger error tolerance (E= 10^{-2}) to maintain the agreement with DI when using scale factors, thus reducing computational time. Good
	Case 1	Case 2	DI
Total CPU time (s)	71483	33169	45520
CPU time for chemistry (s)	33224	2504	9568
Number of queries	2.18×10^7	2.18×10^7	
Primary Retrieves (%)	52.39	87.28	
Secondary Retrieves $(\%)$	18.69	11.13	
Total Retrieves $(\%)$	71.08	98.41	
Replace $(\%)$	19.54	0	
Records	$2.7 imes 10^5$	8014	
Speed-up of chemistry	0.3	3.8	

Table 5.5 A summary of ISAT performance for Cases 1 and 2.

agreement is observed in Fig. 5.8. for void fraction, gas temperature, mass fractions of SiH_4 and H_2 between DI and ISAT.

The performance of ISAT is summarized in Tab. 5.5. For case 1 in which all scale factors are set to unity, the number of records is large since the dependence of reaction rates on temperature generally is exponential. The maximum allowable table size has been exceeded and *replace* occurs. The CPU time for ISAT calculation increases drastically since queries have to be computed by DI and new EOAs are replaced by old EOAs. Case 2 which uses scale factors obtained a speedup of 4 without *replace* and requires less records in the table. The plot of ISAT performance for case 2 is shown in Fig. 5.9. The primary and secondary *retrieves* dominate the queries. After the table has matured, the percentage of *grows* and *adds* reduces greatly.

5.3.2 3D Simulations Using Multiple Tables and Processors

A non-isothermal 3D case was simulated to test ISAT performance with multiple processors and multiple tables. For simplicity of calculation, the computational domain is changed to a rectangular domain with no expansion part. The computational geometry is shown in Fig. 5.10. A Cartesian coordinate system is used for the simulations. Since ISAT generates a separate table on each processor, the performance of different domain decomposition directions are



Figure 5.8 (a) Radial void fraction, (b) gas temperature, (c) mass fraction of SiH_4 and (d) H_2 averaged over 50–100 s and height 2-10 cm using ISAT(DI), ISAT (Case 1) and ISAT (Case 2).



Figure 5.9 Fraction of events versus queries for case 2.

investigated first. The speed-up is tested using the silane case with DI and all cases are run up to 0.2 s. The CPU time per processor for computation is more than 1 hour to make sure that all processors are involved in the computation. The computational domain is decomposed in three ways: (1) parallelization in x direction, (2) parallelization in y direction, (3) parallelization in z direction. The assignments of processors are shown in Tabs. 5.6, 5.7 and 5.8 for each parallelization direction. Speed-up is defined by the formulation

Speed-up
$$(n) = \frac{T_1}{T_n}$$

where Speed-up(n) is the speed-up achieved with n processors, T_1 is CPU time using 1 processor and T_n is CPU time per processor when n processors are used.

Figure 5.11 demonstrates parallelization performance for all of the non-isothermal silane pyrolysis cases. When number of processors is less than or equal to 4, the speed-up with parallelization in different directions are similar. However, the speed-up with parallelization in the y direction is highest (around 3.5) with 8 processors. For the other cases, there are not enough cells on each processor when decomposing the domain in the x and z directions.

The algorithm of ISAT and the properties of fluidized beds should be considered when the domain decomposition direction is chosen. Normally the chemistry is very different along



Figure 5.10 The computational 3D domain for fluidized bed pyrolysis of silane.

the y direction due to different temperatures and species concentrations. The tables on each processor are quite different if the domain is decomposed in the y direction. The table on each processor handles the reaction initial conditions which are smaller than the whole composition space in a fluidized bed reactor. Therefore, the size of tables generated is smaller and CPU time to build these mature tables is less. Each processor tabulates the entire composition space which comprises the dense bed to the freeboard if parallelized in the x or z directions. Thus, parallelization in the x or z directions is not efficient for ISAT. In the following simulations, parallelization in the y direction is chosen and all cases are calculated using 4 processors.

As discussed in §5.3.1, the variables should be scaled by the factors which are the maximum values in the domain. However, the values of variables are quite different in the freeboard than

Table 5.6 Parallelization in the x direction on HPC3.

processors	x	U	z
1	1	1	1
2	2	1	1
4	4	1	1
8	8	1	1

Table 5.7 Parallelization in the y direction on HPC3.

processors	x	y	z
1	1	1	1
2	1	2	1
4	1	4	1
8	1	8	1

Table 5.8 Parallelization in the z direction on HPC3.

processors	x	y	z
1	1	1	1
2	1	1	2
4	1	1	4
8	1	1	8

in the bed. For example, the mass fraction of SiH₄ changes dramatically from the bed to the freeboard, or the solid phase species may not exist in the freeboard. A consequence is that for the processors that handle both freeboard and bed, two tables must be created separately on each processor in order to maintain accuracy and reduce computational time. On every processor, one table is built for chemical reactions in the bed, which include the gas-solid phase reactions and gas phase reactions. Another table is for freeboard and only tabulates the gas phase reactions. Both tables are scaled by different factors which are the maximum values that occurs in the bed and freeboard, respectively. In all simulations, the number of trees is 12 and the error tolerance is 10^{-2} .



Figure 5.11 Speed-up for parallelization in different directions on HPC3 (MPI).

Figure 5.12 show plots of the void fraction, gas temperature and mass fractions of SiH₄ and H₂ in the bed. The plots of gas temperature and mass fractions of SiH₄ in the freeboard are shown in Fig. 5.13. In the 3D simulations, bubbles appear in the center of the domain and solids accumulate near the wall shown in Fig. 5.12(a) unlike the 2D simulations. A high mass fraction of SiH₄ is observed in the center of the bed (Fig. 5.12(c)) and a more uniform distribution of SiH₄ is shown in the freeboard (Fig. 5.13(a)). The mass fraction of H₂ (Fig. 5.12(d)) is almost constant along the radial direction. Gas temperatures are uniform through the whole reactor (Fig. 5.12(b) and Fig 5.13(b)). ISAT(DI) and ISAT compare well for the both regions.

The performance of ISAT for processors 0 and 1 is shown in Tab. 5.9. The domain decomposition and location of tables is shown in Fig. 5.14. Tables 0 and 1 tabulate the chemical reactions in the bed and the freeboard, respectively. As shown in Fig 5.14, table 0 was built on processor 0, and only processor 1 contained both tables 0 and 1. ISAT performance of table 0 and table 1 on processor 1 is shown in Fig. 5.15. For all tables, the total retrieves are more than 99.9%, which indicates that linear interpolation dominates the computation. The speed-up of chemistry is calculated using the CPU time used by ISAT divided by the CPU time used by DI on each processor. For processor 1, the speed-up is around 50. The total CPU time for simulations is reduced by 24% by using ISAT.



Figure 5.12 (a) Radial void fraction, (b) gas temperature, (c) mass fraction of SiH₄ and (d) H₂ averaged over 5–10 s and height 2-10 cm using ISAT(DI), ISAT in the bed.



Figure 5.13 (a) Mass fraction of SiH₄ and (b) gas temperature averaged over 5–10 s and height 20-28 cm using ISAT(DI) and ISAT in the freeboard.

	Processor 0	Proce	ssor 1
	Table 0	Table 0	Table 1
Number of queries	$3.96 imes 10^7$	3.45×10^7	1.28×10^7
Primary Retrieves (%)	63.38	89.48	88.20
Secondary Retrieves (%)	26.53	10.48	11.78
Total Retrieves $(\%)$	99.91	99.96	99.98
Records	859	272	138
Speed-up of chemistry	20.5	48	3.2

Table 5.9 A summary of ISAT performance for 3D simulations.



Figure 5.14 Void fraction at 15 s in 3D simulations using multiple processors and tables.



Figure 5.15 A summary of ISAT performance for (a) the bed (table 0) and (b) the freeboard (table 1) for processor 1.

CHAPTER 6. SIMULATIONS OF FLUIDIZED BED COAL GASIFICATION

The modified MFIX-ISAT algorithm was presented for the isothermal silane pyrolysis case in Section §5.2. The two-dimensional and three-dimensional non-isothermal silane cases were used to investigate the features of ISAT, such as scale variables and multiple tables on multiple processors (§5.3). In this chapter, a coal gasification mechanism is used to test the performance of ISAT with the complex gas-solid reactions using the improved performance parameters discussed in Chapter 5.

6.1 Introduction

Coal is an inhomogeneous organic fuel, formed from partially decomposed and metamorphosed plant materials under high pressures and elevated temperatures. Coals vary greatly in their composition. Coal is the world's most plentiful fossil fuel and also the most abundant fossil fuel in the United States (Smoot, 1993).

Several feasible conversion processes exist such as combustion, gasification and liquefaction (Smoot and Smith, 1986). Gasification of coal is a process in which coal reacts with an oxidizer to produce a gaseous product. The desired products usually include carbon monoxide, hydrogen and methane. The gaseous product can be used as a fuel for generation of electrical power or a synthesis gas. Fluidized beds are widely used for coal gasification due to their advantageous characteristics such as excellent gas-solid contacting and high rate of heat transfer.

The MFIX model of coal chemistry is a modified version of the reaction scheme used by the National Energy Technology Laboratory (Syamlal and Bissett, 1992). The chemical reactions are divided into initial stage reactions, and gasification and combustion reactions. The solid phase consists of coal which contains four pseudo-species: ash, moisture, volatile matter and fixed carbon. The gas phase includes eight species: O_2 , CO, CO_2 , CH_4 , H_2 , H_2O , N_2 and tar. In reactors, ash does not react, and moisture evaporates during the initial stage reactions. Volatile matter releases gas phase species through devolatilization, which depends on the type of coal and the heating process. Fixed carbon takes part in the combustion and gasification reactions. The steps of the chemical reactions for coal are:

1. Initial Stage Reactions

Moisture (coal) \longrightarrow H₂O Volatile Matter \longleftrightarrow Tar + Gases Tar \longrightarrow Fixed Carbon + Gases

2. Gasification Reactions

 $\begin{array}{l} C+H_2O\longleftrightarrow CO+H_2\\ C+CO_2\longleftrightarrow 2CO\\ C+2H_2\longleftrightarrow CH_4 \end{array}$

3. Combustion Reactions

 $\begin{array}{l} 2\mathrm{C} + \mathrm{O}_2 \longrightarrow 2\mathrm{CO} \\ 2\mathrm{CO} + \mathrm{O}_2 \longrightarrow 2\mathrm{CO}_2 \\ \mathrm{CH}_4 + 2\mathrm{O}_2 \longrightarrow \mathrm{CO}_2 + 2\mathrm{H}_2\mathrm{O} \\ 2\mathrm{H}_2 + \mathrm{O}_2 \longrightarrow 2\mathrm{H}_2\mathrm{O} \end{array}$

4. Water Shift Reaction

$$CO + H_2O \longleftrightarrow CO_2 + H_2$$

The coefficients in the initial stage reactions are determined by assuming certain phenomenological rules (Syamlal and Bissett, 1992). The rate expressions for gasification and combustion reactions can be found in Wen et al. (1982).

Proximate Analysis	
Fixed carbon	0.4679
Volatile matter	0.3767
Ash	0.078
Moisture	0.0774
Ultimate Analysis	
С	0.6583
Н	0.0696
Ν	0
0	0.1167
S	0

Table 6.1 Analysis of Illinois No.6 coal.

The feed material used for gasification simulations is Illinois No.6 coal. The feed has a mean particle size of 297 μ m and a density of 2.6 g/cm³. The proximate and ultimate analyses of the coal are given in Tab. 6.1.

6.2 Pairwise Mixing Stirred Reactor

The pairwise mixing stirred reactor (PMSR) model is selected to test the chemical reactions of coal gasification first since the results obtained from the PMSR model provide information for optimizing parameters in ISAT to minimize the computational time for FB reactor simulations. At any time t, the PMSR consists of an even number (N) of particles, the *ith* particle having composition $\phi^{(i)}(t)$ (Pope, 1997; Saxena and Pope, 1999). At t = 0, initial conditions are assigned to all particles in the reactor. Then particles are advanced in discrete time steps, with δt being the specified time step at the discrete times $k\delta t$ (k is an integer that represents time steps). Events occur corresponding to outflow, inflow, and pairing, which cause $\phi^{(i)}(t)$ to change discontinuously. Between discrete times, the composition evolves by a mixing fractional step and a reaction fractional step.

The particles are arranged in pairs; e.g., particles 1 and 2, 3 and 4, ...N - 1 and N are

partners. The mixing fractional step consists of a pair (e.g., p and q) evolving by

$$\frac{\mathrm{d}\phi^{(p)}}{\mathrm{d}t} = -\frac{1}{\tau_{\mathrm{mix}}} \left(\phi^{(p)} - \phi^{(q)}\right) \tag{6.1}$$

$$\frac{\mathrm{d}\phi^{(q)}}{\mathrm{d}t} = -\frac{1}{\tau_{\mathrm{mix}}} \left(\phi^{(q)} - \phi^{(p)}\right) \tag{6.2}$$

where $\tau_{\rm mix}$ is the micromixing time scale.

In the reaction fractional step, each particle evolves according to the chemical source term

$$\frac{\mathrm{d}\phi^{(i)}}{\mathrm{d}t} = S(\phi^{(i)}) \tag{6.3}$$

where S is the source term.

Using $\tau_{\rm res}$ as the specified residence time, inflow and outflow consist of selecting $N\delta t/(2\tau_{\rm res})$ particle pairs at random and replacing their compositions with the inflow compositions. With $\tau_{\rm pair}$ being the specified pairing time scale, $N\delta t/(2\tau_{\rm pair})$ particle pairs (other than inflowing particles) are randomly selected for pairing. These particles and the inflowing particles are then randomly shuffled so that (most likely) they change partners. Equations (6.1) and (6.2) can be solved analytically and need little time for simulations. The total computational time depends on the complexity of the chemical source term and the wide range of chemical time scales in Eqn. (6.3).

The values of the various parameters used in the simulations are shown in Tab. 6.2. As the initial condition, particles in the reactor contain only N_2 and ash with a void fraction of 0.43. The gas and solid temperatures are both 1000 K. The inflow particles include coal with a volumetric fraction 0.002, and air, which has mass fraction of 0.2566 for O_2 and 0.7434 for N_2 . Also, the inlet particles have a temperature of 1000 K. For all simulations using the PMSR model, the Rosenbrock method is used to integrate the ODEs. Simulations were performed on the Sunfire 6800 with ISAT (version 3.0).

The simulations ran up to 1 s, achieving steady state, but the plots only show the results for 0.05 s for sake of clarity. For all ISAT simulations, the variables were scaled by the values in the statistically stationary state. Figure 6.1(a) shows mass fraction of CH_4 and (b) is a

Number of particles	100
Number of time steps, N_{step}	1000
Residence time, $\tau_{\rm res}$	0.001s
Mixing time scale, $\tau_{\rm mix}$	0.0001s
Pairing time scale, τ_{pair}	0.0001s

Table 6.2Parameters used in the PMSR simulations.

Table 6.3 ISAT performance for the PMSR.

	DI	$E = 10^{-4}$	$E = 10^{-3}$	$E = 10^{-2}$
Total CPU time (s)	37606	2625	2151	1834
CPU percentage for chemistry $(\%)$		94.46	93.31	92.07
Number of queries		2.94×10^7	2.94×10^7	2.94×10^7
Retrieves $(\%)$		98.22	98.87	99.04
Records		9.32×10^4	6.22×10^4	4.05×10^4
Speed-up		10.5	17.5	20.5

close-up to compare solutions using DI and solutions for different ISAT error tolerances. ISAT simulations match well with DI for all error tolerances. From the close-up (Fig. 6.1(b)) it can be inferred that ISAT accuracy decreases when the error tolerance increases from 10^{-4} to 10^{-2} . Also, the mass fraction of H₂ and gas temperatures in Fig. 6.2 show that results using ISAT agree well with DI. Thus, the error tolerance (E = 10^{-2}) is reasonably accurate for future simulations. ISAT performance is listed in Tab. 6.3. Chemistry calculations using the PMSR model dominate more than 90% of total CPU time for all cases. The retrieve percentage increases from 98.22 to 99.04 and the number of records reduces by 54% with $E = 10^{-2}$, for a speed-up around 20.5.

6.3 Fluidized Bed Reactor

Figure 6.3 shows the two-dimensional (2D) geometry of a fluidized bed reactor. In the simulations, the solid particles in the bed are ash. A mixture of air and coal enters from the bottom of the bed with velocities of 16.5 cm/s and 0.01 cm/s, respectively. The outlet



Figure 6.1 (a) Mass fraction of CH_4 and (b) the close-up using ISAT(DI) and ISAT.



Figure 6.2 (a) Mass fraction of H_2 and (b) gas temperature using ISAT(DI) and ISAT.



Figure 6.3 Computational domain for coal gasification.

boundary condition is set to a constant pressure of 1.0×10^5 Pa. The temperature inside the bed and at the boundaries is 923 K. No-slip boundary conditions for gas and solid phases are set at the wall. The minimum void fraction in the bed is 0.43 and a Cartesian coordinate system is used for the simulations. The number of cells used in the simulations is 15 and 32 in the radial and axial directions, respectively and simulations are run up to 200 s. All simulations were performed on an Alpha Cluster (HPC3) at Iowa State University with ISAT (version 4.0). For the DI solution of the equations, ODEPACK is used for the calculations. An error tolerance (10^{-2}) is used for ISAT and two tables are built for the bed and freeboard, respectively. All variables are scaled by the maximum values in the reactor obtained by DI.

Figure 6.4 presents void fraction and gas temperature averaged over time using ISAT(DI). The bed expands from 8 to 12 cm during the gasification process. Uniform gas temperature distribution is observed in the bed and higher temperatures occur in the freeboard due to the homogeneous combustion. The averaged contour plots of mass fractions of CO and H₂ using ISAT(DI) are shown in Fig. 6.5. Lower values of mass fractions of CO and H₂ are observed above the distributor plate. Both the mass fractions increase at a higher bed height, where



Figure 6.4 Contour plots of (a) void fraction and (b) gas temperature averaged over 100-200 s using ISAT(DI).



Figure 6.5 Contour plots of mass fractions of (a) CO and (b) H_2 averaged over 100-200 s using ISAT(DI).

coal gasification reactions occur.

Figure 6.6 shows the void fraction, gas temperature and mass fractions of CO and H₂ in the bed. Bubbles appear in the center of the domain and the solids accumulate near the walls (Fig. 6.6(a)). A uniform distribution of gas temperature is observed from Fig. 6.6(b). The plots of mass fractions of CO and H₂ in the freeboard are shown in Fig. 6.7. A high mass fraction of CO is observed in the center of the freeboard (Fig. 6.7(a)) and a more uniform distribution of CO is shown in the bed (Fig. 6.6(c)). Mass fractions of H₂ are uniform for both bed (Fig. 6.6(d)) and freeboard regions (Fig. 6.7(b)). ISAT(DI) and ISAT compare well for both regions.

Table 6.4 presents the performance for ISAT. Tables 0 and 1 tabulate the chemical reactions in the bed and the freeboard, respectively. ISAT performance of table 0 and table 1 is shown in Fig. 6.8. For all tables, the total retrieves are more than 99.9%, which indicates that the tables are mature. The speed-up of chemistry is around 16 for 200 simulation seconds. The total CPU time for MFIX-ISAT simulations is reduced by 10% by using ISAT.



Figure 6.6 (a) Void fraction, (b) gas temperature, (c) mass fractions of CO and (d) H_2 averaged over 100–200 s and height 0-4 cm using ISAT(DI) and ISAT in the bed.



Figure 6.7 (a) Mass fractions of CO and (b) H_2 averaged over 100–200 s and height 15-25 cm using ISAT(DI) and ISAT in the freeboard.

Table 6.4ISAT performance for fluidized bed gasification.

	Table 0	Table 1
Number of queries	4.12×10^7	5.33×10^7
Primary Retrieves (%)	82.35	93.05
Secondary Retrieves (%)	17.61	6.94
Total Retrieves $(\%)$	99.96	99.99
Records	1801	42
Speed-up of chemistry	16.1	



Figure 6.8 A summary of ISAT performance for (a) the bed (table 0) and (b) the freeboard (table 1).

CHAPTER 7. EFFECTS OF USING TWO- VERSUS THREE-DIMENSIONAL COMPUTATIONAL MODELING OF FLUIDIZED BEDS

CFD simulations of fluidized beds using an Eulerian-Eulerian model are computationally expensive. In order to simulate phenomena such as particle segregation and elutriation, multiple types of particles should be considered. Each particle type has a unique diameter with identical material properties. Thus, three-dimensional simulations of fluidized beds are usually prohibitive. Two-dimensional simulations are commonly used to compare with experiments. This chapter focuses on the use of coordinate systems and configuration to model fluidized bed reactors. Three different fluidization regimes, bubbling, slugging and turbulent regimes, are investigated with inlet gas velocities, $1.25U_{\rm mf}$, $1.86U_{\rm mf}$ and $4.04U_{\rm mf}$, respectively. The individual terms of the governing equations in two-dimensional (2D) and three-dimensional (3D) simulations with the cylindrical and Cartesian coordinate systems are evaluated. The additional terms appearing in 3D simulations are used to explain the discrepancy between 2D and 3D simulations.

7.1 Introduction

Many numerical simulations of dense fluidized beds based on the Eulerian-Eulerian model have been performed. In an early summary paper by Enwald et al. (1996), most published literature presented 2D simulations since full 3D simulations were computationally prohibitive due to memory and processor speeds. To date, even with improved computational facilities, 3D simulations are still expensive as the equations have to be integrated for long times with small time-steps to compute the inherently chaotic and transient fluidization processes. According to Geldart (1986), there are different coalescence effects in 2D and 3D beds. Also, smaller bubbles (Geldart, 1970) and a lower rise velocity (van Wachem et al., 1998) are usually observed with 2D beds. Therefore, one question that persists is the validity of using 2D simulations to represent the true dynamical behavior of fluidized beds. Another question is the choice of coordinate system and configuration used to model a cylindrical reactor, which is the most common geometry used in industry.

Comparison between simulations and experiments is a common way to validate CFD models. For example, the 2D simulations of rectangular fluidized beds using the Cartesian coordinate system by Johansson et al. (2006) were compared with experiments, and the general trends indicated that 2D simulations show good agreement. The 2D Cartesian coordinate system usually is used to simulate 3D rectangular fluidized beds, especially for pseudo-2D experiments. Typically, pseudo-2D beds have a small depth compared to the height and width. Thus the particle motion is suppressed in the depth direction. Many researchers have applied 2D Cartesian simulations to model pseudo-2D beds (Sun and Battaglia, 2006; Chiesa et al., 2005; Goldschmidt et al., 2001, 2004; Boemer et al., 1998). Johansson et al. (2006) and Enwald et al. (1999) also employed a 2D Cartesian system to represent 3D rectangular geometries which were not considered as pseudo-2D beds.

Both the 2D cylindrical, which assumes symmetry in the azimuthal direction, and 2D Cartesian coordinate systems are employed for simulations of a cylindrical fluidized bed. Syamlal and O'Brien (2003) applied the 2D cylindrical coordinate system for catalytic decomposition of ozone in a fluidized bed reactor. Their simulations showed that bubbles formed near the wall and migrated toward the centerline. However, bubbles did not reach the center of the bed. The atypical flow dynamics may be due to the axisymmetric assumption which is not representative of a real fluidized bed. An alternative approach is to simulate a cylindrical fluidized bed using the 2D Cartesian coordinate system, which represents an axial slice at the center of the reactor. Such simulations include work by Hulme et al. (2005), Chandrasekaran et al. (2005), Zimmermann and Taghipour (2005), McKeen and Pugsley (2003), Pain et al. (2002) and van Wachem et al. (1999). However, there are few articles in the literature that discuss the justification for using 2D simulations for fluidized beds.

Peirano et al. (2001) performed numerical simulations of a rectangular bubbling fluidized bed using the code ESTET/ASTRID. Numerical results were compared to experimental values for the power spectra of pressure fluctuations, bed height, and probability density function of particle volume fraction. They observed a significant difference between 2D and 3D simulations, and only 3D simulations could predict the correct bed height and pressure spectra. Cammarata et al. (2003) also validated the bubbling behavior predicted by 2D and 3D simulations for a rectangular fluidized bed using commercial software, CFX. The bed expansion, bubble hold-up and bubble size calculated from 2D and 3D simulations were compared with the predictions obtained from the Darton equation (Darton et al., 1977). A more realistic physical behavior of fluidization was obtained using 3D simulations. Both Peirano et al. (2001) and Cammarata et al. (2003) concluded that 3D simulations should be preferably performed except maybe in cases where the flow is by nature two dimensional. However, these authors indicate that 2D simulations could be used to conduct sensitivity analyses. For a cylindrical fluidized bed, there are no studies comparing 2D and 3D simulations.

Briongos and Guardiola (2005) presented a new method to scale hydrodynamic data between a 2D and 3D fluidized bed applying the chaos scale-up methodology. They tested the methodology for 2D fluidized beds and 3D cylindrical fluidized beds. The findings suggested that 2D and 3D fluidized beds are dynamically similar if the fluidized beds have the same flow properties, which is represented by the Kolmogorov entropy under the same fluidization conditions. However, the results presented were limited to small ranges of gas velocities and particle properties.

7.2 Hydrodynamics

The aim of the present work is to show the range of validity for employing simulations based on a 2D Cartesian coordinate system to approximate both cylindrical and rectangular fluidized beds. The simulations were conducted for three different inlet gas velocities, $1.25U_{\rm mf}$, $1.86U_{\rm mf}$ and $4.04U_{\rm mf}$ ($U_{\rm mf}$ is the minimum fluidization velocity) which correspond to the bubbling,



Figure 7.1 Computational domains for a (a) cylindrical and (b) rectangular fluidized bed reactor.

slugging and turbulent regimes of fluidized beds. For bubbling fluidized beds, the results of 2D and 3D simulations are presented for both cylindrical and rectangular domains. The comparison of 2D and 3D simulations for a cylindrical geometry is shown for the slugging and turbulent regimes. MFIX will be used for all simulations; for details, refer to Chapter 3.

7.2.1 Domain Discretization and Grid Resolution Study

The computational domains for the cylindrical and rectangular fluidized beds are shown in Fig. 7.1. The heights of the domains are 100 cm for all the simulations. The diameter of the cylinder is 14 cm and the cross section of the rectangular geometry is a square with a length of 14 cm per side. Three dimensional simulations of the cylindrical and rectangular domains were calculated using the cylindrical and Cartesian coordinate systems, respectively. For 2D simulations of the cylindrical domain, a 2D cylindrical (axisymmetric) coordinate system was used with a 7 cm radius. A two-dimensional Cartesian coordinate system was considered, representing a plane for both the cylindrical and rectangular domains with a width of 14 cm.

Finite volume schemes were used to discretize the governing equations. The SIMPLE algorithm was modified for multiphase flow to solve the algebraic equations (Syamlal, 1998). In simulations, the Superbee method was used as spatial discretization. A PDE for the granular temperature was solved. The walls were modeled using no-slip boundary conditions for both gas and solid phases. The free-slip boundary condition was used at the centerline for the 2D cylindrical domain. A uniform superficial gas velocity was employed at the bottom of the bed and ambient pressure was specified at the exit. Three superficial gas velocities were considered, representing three different flow regimes: a bubbling bed $(1.25U_{\rm mf})$, a slugging bed $(1.86U_{\rm mf})$ and a turbulent bed $(4.04U_{\rm mf})$. For the initial condition, the fluidized bed was at minimum fluidization with a solid volume fraction of 0.58. The settled bed height was 50 cm for the bubbling and slugging regimes and 30 cm for the turbulent regime. The freeboard was considered to be occupied by gas only. The solid particles have a density of 2.0 g/cm^3 , diameter of 0.04 cm and are classified as a Geldart group B material with an assumed $U_{\rm mf}$ of 21.56 cm/s. Neither the choice of the particles nor the drag formulation would necessarily alter the conclusions (as long as the models and parameters are fixed for the different cases) of the paper as the focus is mainly on understanding the differences in the choice of coordinate systems. The particles were fluidized by air at ambient conditions.

Two dimensional simulations using the cylindrical coordinate system were used to determine grid independence with a superficial gas velocity of 40 cm/s. Beginning with 7 and 100 cells in the radial and axial directions, respectively, the number of cells was doubled, as shown in Tab. 7.1. Figure 7.2 shows the profiles of the void fraction versus radial distance averaged over time (10–20 s) for two fixed bed heights. There is some change in the profiles when the grid is refined from 7×100 to 14×200 . However, further doubling the grid did

Grid	$\triangle x (cm)$	$\triangle y (cm)$
7×100	1.0	1.0
14×200	0.5	0.5
28×400	0.25	0.25
56×800	0.125	0.125

Table 7.1 The grid resolution and cell sizes for 2D cylindrical simulations.



Figure 7.2 Void fraction versus radial distance for different grids at (a) 20 cm and (b) 50 cm above the inlet for a superficial gas velocity of 40 cm/s.

not result in significant differences in the solutions. This also corresponds to the resolution of 10 times the particle diameter which seems to be the typical resolution required to have grid independent simulations (Guenther and Syamlal, 2001). As a compromise between accuracy and computational cost, a uniform grid of 14×200 ($5 \times 5 \text{ mm}^2$ cells) was employed in all the simulations.

A further study examined 2D and 3D simulations using the cylindrical coordinate system for a superficial gas velocity of 40 cm/s. A summary of the grid resolutions is found in Tab. 7.2. The void fraction versus radial distance at 20 cm above the inlet is shown in Fig. 7.3. Unphysical results were obtained when using the 2D cylindrical coordinate system, as the

	Grid
2D cylindrical	14×200
3D cylindrical	$14\times 200\times 2$
3D cylindrical	$14\times 200\times 3$
3D cylindrical	$14\times 200\times 12$
2D Cartesian	28×200
3D Cartesian	$28\times200\times28$

Table 7.2 The grid resolution for the cylindrical and Cartesian systems.

bubbles were repelled by the centerline free-slip boundary condition and the bubbles coalesced away from the centerline, indicative of higher void fraction. Next, the 3D cylindrical domain with 2 cells in the azimuthal direction was used to represent the axisymmetric condition, however, lower void fraction was also observed near the centerline. Interestingly, increasing the number of azimuthal cells to 3 captured the correct trend, allowing bubbles to rise along the centerline (Guenther et al., 2001). Also, a 3D cylindrical calculation with 12 azimuthal cells was tested, which reached the upper limit of the computational resources in both memory and time on an Alpha cluster (DEC Alpha EV6.7 processor running at 833 MHz). It should be noted that using 12 cells to discretize the azimuthal direction may be considered a coarse resolution, effectively producing an axisymmetric flow. According to Guenther et al. (2001), azimuthal cell discretization of 24 or greater were considered grid independent solutions. However, they showed that 12 cells in the azimuthal direction were qualitatively similar to 24 and 36 cells. For this study, only results for the 3D cylindrical system with 3 and 12 cells in the azimuthal direction will be used in the following discussions.

7.2.2 Blending Formulation for Kinetic-Frictional Transition

Continuum simulations using Eulerian-Eulerian formulations like MFIX have only an approximate description of the stresses arising from the granular material. This is due to the fact that granular materials behave differently when they are flowing rapidly as compared to when they are flowing slowly with longer-lasting contacts. The behavior of rapidly flowing



Figure 7.3 Void fraction versus radial distance to compare 2D and 3D cylindrical grids.

granular materials, where the particle-particle interactions are largely dominated by binary collisions, is described quite well by the kinetic theory of granular materials and has been employed in MFIX (e.g. Lun et al. (1984)). The same theory cannot be applied for slowly moving or static assemblies of granular materials, where longer-lasting contacts between particles transmit forces over relatively long-range stress chains. There are several continuum models which approximate the behavior in this regime, and the one used in MFIX is the Schaeffer model (Schaeffer, 1987). The rheological behavior of the transition zone between these two granular regimes is poorly understood (e.g. Savage (1998)) and currently not employed in the kinetic theory—plastic formulation in the default MFIX model.

However, many important gas-solid flow applications (including bubbling beds, spouted beds etc.) have rapid transitioning between these two regimes at various locations in the bed. In the current MFIX implementation, the granular stresses transition from the rapidly shearing regime to quasi-static assemblies for void fractions below an ad-hoc critical packing void fraction (ϵ^*); this is depicted in Fig. 7.4. This step transitioning (discontinuity) not only causes numerical instabilities leading to slow convergence but is also found to generate spurious solutions in spouted beds. In order to address these problems, an approximate transitioning function, referred to here as the blending stress function, was constructed to provide a smooth



Figure 7.4 A cartoon depicting the transition from the rapidly shearing viscous regime to the slow shearing plastic regime.

transition between the two granular regimes in the void-fraction space. To be more rigorous, this function should be both a function of local void fraction and shear rate, however, the results to date have been very promising for the case where the blending function is dependent only on void fraction. Thus this simpler formulation is used in this work. Here a brief description of the blending stress formulation is presented.

The normal stress (pressure) in the conventional granular stress model is given as:

$$P_{\rm granular} = \begin{cases} P_{\rm critical} = 10^{25} \left(\epsilon^* - \epsilon_{\rm g}\right)^{10} & {\rm for} \ \epsilon_{\rm g} < \epsilon^* \\ \\ P_{\rm kinetic \ theory} & {\rm for} \ \epsilon_{\rm g} \geq \epsilon^* \end{cases}$$

The viscous stresses and granular conductivity also switch between the critical state theory and kinetic theory similar to the granular pressure. A blending function is introduced in the void-fraction space with the following properties:

- a smooth but rapid transitioning around the critical packing void fraction (ϵ^*)
- approaches zero at $\epsilon^*-\delta\epsilon$
- approaches unity at $\epsilon^* + \delta \epsilon$

Some obvious choices for this function include using a hyperbolic tangent (used in many gridstretching programs, blending drag formulations etc.) or a sigmoidal function (used for rapid transitioning, e.g., for combustion efficiency dependency on equivalence ratio, as in Daw et al. (1996)). Both of these functions for blending the granular stresses have been employed and a hyperbolic tangent function has been used in this work because of its simplicity. However, for details of how these two different functions perform for a bin discharge example, refer to Pannala et al. (2006).

The hyperbolic tangent blending function is given as:

$$\phi\left(\epsilon_{\rm g}\right) = \left[\tanh\left(\frac{2\pi\left(\epsilon - \epsilon^*\right)}{\epsilon_u - \epsilon_l}\right) + 1 \right] / 2 \tag{7.1}$$

where $\phi(\epsilon_l) = 0.0$ and $\phi(\epsilon_u) = 1.0$; $\epsilon_l = 0.99\epsilon^*$ and $\epsilon_u = 1.01\epsilon^*$. Figure 7.5 is a plot of the hyperbolic tangent blending function shown versus void fraction, ϵ_g . The parameters ϵ_l and ϵ_u are equal to $\epsilon^* \pm \delta \epsilon$. Once the blending function is constructed, the granular stresses are blended in the following manner:

$$P_{\text{granular}} = \begin{cases} \phi\left(\epsilon_{\text{g}}\right) P_{\text{kinetic theory}} + \left[1 - \phi\left(\epsilon_{\text{g}}\right)\right] 10^{25} \left(\epsilon_{u} - \epsilon_{\text{g}}\right)^{10} & \text{for } \epsilon_{\text{g}} < \epsilon_{u} \\ P_{\text{kinetic theory}} & \text{for } \epsilon_{\text{g}} \ge \epsilon_{u} \end{cases}$$

Shear stress and the granular conductivity are also blended using the same function.

The influence of the blending function on accuracy and computational time was tested with a 2D Cartesian coordinate system. Figure 7.6 presents the bed height versus the mean void



Figure 7.5 Hyperbolic tangent blending function (in blue) and its application to blending granular stresses around the critical state void fraction.

fraction across the bed with and without the blending function. Higher bed height is obtained for the case using the blending function. The void fraction and gas and solid velocities at two different heights show a similar trend for both cases, as shown in Fig. 7.7. It was found that the blending formulation improves the accuracy in the bin discharge flow (Pannala et al., 2006). In addition, the smooth transition between the two granular flow regimes improves the scaling characteristics of the computations and significantly improves the convergence characteristics, especially in 3D cylindrical coordinates. For the above reasons, the blending formulation for transitional flows is used in all further simulations in this study.



Figure 7.6 Void fraction versus bed height at a superficial gas velocity of 40 cm/s with and without the blending function.


Figure 7.7 Void fraction and gas and solid velocities versus radial distance at 20 cm (left) and 50 cm (right) above the inlet for a superficial gas velocity of 40 cm/s with and without the blending function.

7.2.3 Results and Discussion

The results are presented in three parts. In each part, the differences between 2D and 3D simulations are shown for each fluidized regime. Three coordinate systems are examined for the bubbling regime: 2D Cartesian, 3D Cartesian and 3D cylindrical. Table 7.2 summaries the grid resolutions and corresponding coordinate systems. A goal of the comparisons is to determine if a 2D Cartesian representation is reasonable for either the 3D Cartesian or cylindrical reactor domains (see Fig. 7.1). For the slugging and turbulent regimes, the 3D Cartesian domain is not considered.

7.2.3.1 Bubbling Fluidization

According to experimental evidence, Geldart group B material should exhibit a bubbling behavior when the gas velocity exceeds the minimum fluidization velocity $(U_{\rm mf})$. In this study, the gas-solid drag coefficient by Syamlal et al. (1993) was used in the simulations. The drag model parameters have been adjusted to achieve the minimum fluidization velocity 21.56 cm/s. Therefore the fluidization velocity was chosen as 27 cm/s (~1.25 $U_{\rm mf}$) to simulate the bubbling regime and used in the following simulations.

A time sequence of void fraction using a 2D Cartesian system is shown in Fig. 7.8. Small bubbles develop near the inlet and travel through the bed, forming larger bubbles by coalescence. Figure 7.9 presents mean void fraction varying along the bed height averaged over 10–20 s for the four cases: 2D Cartesian, 2D axisymmetric, 3D cylindrical and 3D Cartesian. Results indicate that similar bed heights are obtained for all four test cases. The time-averaged void fraction, and gas and solid velocities at heights of 20 and 40 cm are shown in Fig. 7.10. The results using a 3D cylindrical system are averaged over all planes in the azimuthal direction. The 3D Cartesian results are averaged over the vertical planes. The results for the 2D Cartesian system agree well with results using the 3D cylindrical system with 12 cells for both void fraction, and gas and solid velocities. However, at the lower bed height (20 cm) bubbles stay away from the centerline with the 3D cylindrical system with 3 cells in the azimuthal direction. The results with the 3D Cartesian system show slightly lower gas and solid velocities than the 2D Cartesian system due to the wall effect in the third direction.

7.2.3.2 Slugging Fluidization

In general, slugging fluidization can occur in beds of small diameter if three conditions are met: 1) maximum bubble size is greater than 0.6 times the diameter of the bed; 2) the superficial gas velocity is sufficiently high; 3) the bed is sufficiently deep. Geldart (1986) proposed a criterion for slugging flows:

$$U_{\rm ms} = U_{\rm mf} + 1.6 \times 10^{-3} (60D^{0.175} - H_{\rm mf})^2 + 0.07(g100D)^{0.5}$$
(7.2)

where D is the bed diameter, $H_{\rm mf}$ is the bed height at minimum fluidization and g is gravity. The minimum slugging velocities $(U_{\rm ms})$ computed using Eqn. (7.2) for this case is 33.03 cm/s $(1.53U_{\rm mf})$. A superficial gas velocity 40 cm/s (~1.86 $U_{\rm mf}$) is chosen to simulate the slugging regime.

Instantaneous plots of void fraction are shown in Fig. 7.11. Large bubbles appear in the bed and grow by coalescence until they reach the width of the bed. Figure 7.12 shows mean void fraction varying along the bed height averaged over 10–20 s. The 2D Cartesian system shows a slightly higher bed expansion than both cases using the 3D cylindrical grid resolutions. The averaged void fractions, and gas and solid velocities at heights of 20 and 50 cm are shown in Fig. 7.13. The results of void fraction using the 2D Cartesian system are in very good agreement with results using the 3D cylindrical system with 12 azimuthal cells. However, there are discrepancies for gas velocities in the bed. For gas and solid velocities, the 3D cylindrical system with 3 azimuthal cells obtained correct trends (Guenther et al., 2001) and compared better than the 2D Cartesian system with the 3D cylindrical system using 12 cells.

7.2.3.3 Turbulent Fluidization

The transition to turbulent fluidization is associated with many factors, such as fluidization pressure, particle properties and the geometric conditions of the bed. Based on the experimental results of the transition to turbulent regime shown by Yang (2003), a gas velocity of 87 cm/s ($\sim 4.04U_{\rm mf}$) is chosen for the simulations. The initial bed height is reduced to 30 cm compared to previous simulations in order to prevent the bed material from being elutriated.

Instantaneous plots of void fraction are shown in Fig. 7.14. A more rapid and irregular development of bubble coalescence is observed, forming very large bubbles. The bed surface fluctuates due to the bursting of bubbles which also can be observed from Fig. 7.15.

The mean void fraction is not uniform upward through the bed as with the other two regimes (refer to Figs. 7.9 and 7.12). The bursting of bubbles is demonstrated with the increasing void fraction over the bed height of 30–60 cm. The time-averaged void fraction, and gas and solid velocities at 20 and 50 cm are shown in Fig. 7.16. Predictions of void fraction agree very well at lower bed heights, but there are discrepancies higher in the bed where the flow is more turbulent. The gas and solid velocities are in reasonable agreement. Overall, the 2D Cartesian simulations under-predicted the velocities.

7.2.4 Summary

The computational time required for each case representing the cylindrical domain and various flow regimes is listed in Tab. 7.3. As expected, the 3D cylindrical simulation takes more time per computation, even with as few as 3 cells in the azimuthal direction. The difference in gas velocities between 2D Cartesian and 3D cylindrical with each case increases with increasing inlet gas velocities. For bubbling fluidization, satisfactory qualitative agreement between 2D and 3D simulations is observed for both cylindrical and rectangular fluidized beds. The 2D Cartesian system can predict the correct multiphase fluid dynamics with much less computational time. These results indicate that a 2D Cartesian system can be used to successfully simulate and predict a bubbling regime. However, caution must be exercised when using 2D Cartesian coordinates for other fluidized regimes.



Figure 7.8 Contours of instantaneous void fraction at a superficial gas velocity of 27 cm/s for a bubbling fluidized bed with 2D Cartesian simulations.



Figure 7.9 Time averaged void fraction versus bed height at a superficial gas velocity of 27 cm/s for a bubbling fluidized bed comparing coordinate systems.



Figure 7.10 Void fraction and gas and solid velocities versus radial distance at 20 cm (left) and 40 cm (right) above the inlet for a superficial gas velocity of 27 cm/s for a bubbling fluidized bed.



Figure 7.11 Contours of instantaneous void fraction at a superficial gas velocity of 40 cm/s for a slugging fluidized bed with 2D Cartesian simulations.



Figure 7.12 Time averaged void fraction versus bed height at a superficial gas velocity of 40 cm/s for a slugging fluidized bed comparing coordinate systems.



Figure 7.13 Void fraction and gas and solid velocities versus radial distance at 20 cm (left) and 50 cm (right) above the inlet for a superficial gas velocity of 40 cm/s for a slugging fluidized bed.



Figure 7.14 Contours of instantaneous void fraction at a superficial gas velocity of 87 cm/s for a turbulent fluidized bed with 2D Cartesian simulations.



Figure 7.15 Time averaged void fraction versus bed height at a superficial gas velocity of 87 cm/s for a turbulent fluidized bed comparing coordinate systems.



Figure 7.16 Void fraction and gas and solid velocities versus radial distance at 20 cm (left) and 50 cm (right) above the inlet for a superficial gas velocity of 87 cm/s for a turbulent fluidized bed.

	3D cylindrical	3D cylindrical	2D Cartesian
Grid	$14\times 200\times 3$	$14\times 200\times 12$	28×200
CPU time (h) (27 cm/s)	114.8	616.8	49.6
CPU time (h) (40 cm/s)	176.0	1118.4	64.4
CPU time (h) (87 cm/s)	348.4	1393.6	79.6

 Table 7.3
 Computational time for simulations.

7.3 Analyses

To get a better insight into the complex phenomena associated with fluidized beds, an analysis has been performed to explain the discrepancies observed between the 2D and 3D simulations. The governing equations are compared for each coordinate system to isolate the terms, and hence the physics, that are not similar. Results are presented in two sections. First, the difference in simulations using 2D Cartesian and 3D cylindrical coordinate systems with three superficial gas velocities is presented. Then the difference between 2D and 3D Cartesian simulations with the superficial gas velocity $1.25U_{\rm mf}$ is shown.

7.3.1 2D Cartesian and 3D Cylindrical Coordinates

The governing equations and constitutive relations of MFIX are explained in detail in Chapter 3. The vector form of the gas momentum equation is repeated here:

$$\frac{\partial}{\partial t} \left(\epsilon_{\rm g} \rho_{\rm g} \vec{V}_{\rm g} \right) + \nabla \cdot \left(\epsilon_{\rm g} \rho_{\rm g} \vec{V}_{\rm g} \vec{V}_{\rm g} \right) = \nabla \cdot \overline{\overline{S}}_{\rm g} + \epsilon_{\rm g} \rho_{\rm g} \vec{g} - \vec{I}_{\rm gs}$$
(7.3)

where the subscript g indicates the gas phase. Equation (7.3) can be expressed for a 2D Cartesian and 3D cylindrical coordinate system, as shown in Fig. 7.17 with the corresponding velocity components. The expanded form of the 2D Cartesian system for the streamwise

direction is:

$$\frac{\partial}{\partial t} \left(\epsilon_{\rm g} \rho_{\rm g} v_{\rm g} \right) + \frac{\partial}{\partial x} \left(\epsilon_{\rm g} \rho_{\rm g} u_{\rm g} v_{\rm g} \right) + \frac{\partial}{\partial y} \left(\epsilon_{\rm g} \rho_{\rm g} v_{\rm g} v_{\rm g} \right) = -\frac{\partial P_{\rm g}}{\partial y} + \frac{\partial}{\partial x} \left[\epsilon_{\rm g} \mu_{\rm g} \left(\frac{\partial u_{\rm g}}{\partial y} + \frac{\partial v_{\rm g}}{\partial x} \right) \right]$$

$$+ \frac{\partial}{\partial y} \left(2\epsilon_{\rm g} \mu_{\rm g} \frac{\partial v_{\rm g}}{\partial y} \right) - \frac{\partial}{\partial y} \left[\frac{2}{3} \epsilon_{\rm g} \mu_{\rm g} \left(\frac{\partial u_{\rm g}}{\partial x} + \frac{\partial v_{\rm g}}{\partial y} \right) \right] + F_{\rm gs} \left(v_{\rm s} - v_{\rm g} \right) - \epsilon_{\rm g} \rho_{\rm g} g$$

$$(7.4)$$

where v and u are the velocity components in the streamwise (y) and cross-stream (x) directions, respectively.

The 3D cylindrical coordinate system is

$$\frac{\partial}{\partial t} \left(\epsilon_{g} \rho_{g} v_{g} \right) + \frac{\partial}{\partial r} \left(\epsilon_{g} \rho_{g} u_{g} v_{g} \right) + \frac{\epsilon_{g} \rho_{g} u_{g} v_{g}}{r} + \frac{\partial}{\partial y} \left(\epsilon_{g} \rho_{g} v_{g} v_{g} \right) + \frac{1}{r} \frac{\partial}{\partial \theta} \left(\epsilon_{g} \rho_{g} v_{g} w_{g} \right) = -\frac{\partial P_{g}}{\partial y} \quad (7.5)$$

$$+ \frac{\partial}{\partial r} \left[\epsilon_{g} \mu_{g} \left(\frac{\partial u_{g}}{\partial y} + \frac{\partial v_{g}}{\partial r} \right) \right] + \frac{\epsilon_{g} \mu_{g}}{r} \left(\frac{\partial u_{g}}{\partial y} + \frac{\partial v_{g}}{\partial r} \right) + \frac{\partial}{\partial y} \left(2 \epsilon_{g} \mu_{g} \frac{\partial v_{g}}{\partial y} \right)$$

$$+ \frac{1}{r} \frac{\partial}{\partial \theta} \left[\epsilon_{g} \mu_{g} \left(\frac{1}{r} \frac{\partial v_{g}}{\partial \theta} + \frac{\partial w_{g}}{\partial y} \right) \right] - \frac{\partial}{\partial y} \left[\frac{2}{3} \epsilon_{g} \mu_{g} \left(\frac{\partial u_{g}}{\partial r} + \frac{\partial v_{g}}{\partial y} + \frac{1}{r} \frac{\partial w_{g}}{\partial \theta} + \frac{u_{g}}{r} \right) \right]$$

$$+ F_{gs} \left(v_{s} - v_{g} \right) - \epsilon_{g} \rho_{g} g$$

where u, v and w are three components of gas velocity, in the radial (r), streamwise (y) and azimuthal (θ) directions. As shown in Fig. 7.17, x and y in the 2D Cartesian coordinate system correspond to r and y in the 3D cylindrical coordinate system, respectively. Thus, two additional terms appear on the left side of Eqn. (7.5), namely $\frac{\epsilon_g \rho_g u_g v_g}{r}$ and $\frac{1}{r} \frac{\partial}{\partial \theta} (\epsilon_g \rho_g v_g w_g)$. There are three additional terms on the right side of Eqn. (7.5): $\frac{\epsilon_g \mu_g}{r} \left(\frac{\partial u_g}{\partial y} + \frac{\partial v_g}{\partial r}\right), \frac{1}{r} \frac{\partial}{\partial \theta} \left[\epsilon_g \mu_g \left(\frac{1}{r} \frac{\partial v_g}{\partial \theta} + \frac{\partial w_g}{\partial y}\right)\right]$ and $-\frac{\partial}{\partial y} \left[\frac{2}{3} \epsilon_g \mu_g \left(\frac{1}{r} \frac{\partial w_g}{\partial \theta} + \frac{u_g}{r}\right)\right]$. Other terms are the common terms which exist in both Eqs. (7.4) and (7.5), such as $\frac{\partial}{\partial t} (\epsilon_g \rho_g v_g), -\frac{\partial P_g}{\partial y}$ and so on. Refer to Tab. 7.4 for a summary of the terms to be compared.

The contour plots of gas velocity in three directions for different regimes are shown in Figs. 7.18, 7.19 and 7.20. All three velocities increase as the inlet gas velocities increase and the axial direction with the velocity $v_{\rm g}$ is the dominant direction. To appreciate the physical behavior of every term appearing in Eqs. (7.4) and (7.5) relative to the solution of $v_{\rm g}$, a comparison of these terms is pursued. In order to compare the magnitude of all terms, finite differencing is used to discretize the partial derivatives. For example, three adjacent planes



Figure 7.17 Domains for a (a) 3D cylindrical and (b) 2D Cartesian coordinate system and the corresponding velocity components.



Figure 7.18 The contour plots of gas velocities in (a) r- (b) y- and (c) $\theta-$ directions for a superficial gas velocity $1.25U_{\rm mf}$ averaged over 10–20 seconds with 3D cylindrical simulations.



Figure 7.19 The contour plots of gas velocities in (a) r- (b) y- and (c) $\theta-$ directions for a superficial gas velocity $1.86U_{\rm mf}$ averaged over 10–20 seconds with 3D cylindrical simulations.



Figure 7.20 The contour plots of gas velocities in (a) r- (b) y- and (c) $\theta-$ directions for a superficial gas velocity $4.04U_{\rm mf}$ averaged over 10–20 seconds with 3D cylindrical simulations.

terms	2D Cartesian	3D cylindrical	3D Cartesian
(1)		$rac{\epsilon_{ m g} ho_{ m g}u_{ m g}v_{ m g}}{r}$	
(2)		$rac{1}{r}rac{\partial}{\partial heta}\left(\epsilon_{ m g} ho_{ m g}v_{ m g}w_{ m g} ight)$	
(3)		$rac{\epsilon_{ m g}\mu_{ m g}}{r}\left(rac{\partial u_{ m g}}{\partial y}+rac{\partial v_{ m g}}{\partial r} ight)$	
(4)		$\frac{1}{r}\frac{\partial}{\partial\theta}\left[\epsilon_{\rm g}\mu_{\rm g}\left(\frac{1}{r}\frac{\partial v_{\rm g}}{\partial\theta}+\frac{\partial w_{\rm g}}{\partial y}\right)\right]$	
(5)		$\frac{\partial}{\partial y} \left[\frac{1}{2} \epsilon_{\rm g} \mu_{\rm g} \left(\frac{1}{r} \frac{\partial w_{\rm g}}{\partial \theta} + \frac{u_{\rm g}}{r} \right) \right]$	
(6)	$-\frac{\partial P_{\rm g}}{\partial y}$	$-\frac{\partial P_{\rm g}}{\partial y}$	$-\frac{\partial P_{\rm g}}{\partial y}$
(7)	$\frac{\partial}{\partial t} \left(\epsilon_{\mathrm{g}} \rho_{\mathrm{g}} v_{\mathrm{g}} \right)$	$rac{\partial}{\partial t}\left(\epsilon_{ m g} ho_{ m g}v_{ m g} ight)$	$rac{\partial}{\partial t}\left(\epsilon_{ m g} ho_{ m g}v_{ m g} ight)$
(8)	$F_{\rm gs}\left(v_{\rm s}-v_{\rm g} ight)$	$F_{ m gs}\left(v_{ m s}-v_{ m g} ight)$	$F_{\rm gs}\left(v_{\rm s}-v_{\rm g}\right)$

Table 7.4 A summary of the terms to be compared with different coordinate systems.

k - 1, k and k + 1, as shown in Fig. 7.21, are randomly chosen to calculate the values of the terms in Eqn. (7.5) since the flow patterns in a cylinder are usually symmetric. For the first order derivatives such as $\frac{1}{r}\frac{\partial}{\partial\theta}(\epsilon_{\rm g}\rho_{\rm g}v_{\rm g}w_{\rm g})$, forward differencing is used for the k and k + 1planes. Central differencing is applied to calculate the second order derivatives, such as the term $\frac{\partial}{\partial y}\left[\frac{2}{3}\epsilon_{\rm g}\mu_{\rm g}\left(\frac{1}{r}\frac{\partial w_{\rm g}}{\partial\theta}+\frac{u_{\rm g}}{r}\right)\right]$. The k plane is used to compute the term $\frac{\epsilon_{\rm g}\rho_{\rm g}u_{\rm g}v_{\rm g}}{r}$.

To evaluate the importance of the additional terms appearing in the 3D cylindrical system, their numerical values are averaged over 10–20 seconds. Figures 7.22, 7.23 and 7.24 show the contour plots of the additional terms for three different superficial gas velocities, $1.25U_{\rm mf}$, $1.86U_{\rm mf}$ and $4.04U_{\rm mf}$, respectively, for the 3D cylindrical simulations. The values of all five additional terms increase as the superficial gas velocity increases. The terms of (1) $\frac{\epsilon_{\rm g}\rho_{\rm g}u_{\rm g}v_{\rm g}}{r}$ and (2) $\frac{1}{r}\frac{\partial}{\partial\theta}\left(\epsilon_{\rm g}\rho_{\rm g}v_{\rm g}w_{\rm g}\right)$ have increased in a proportion greater than terms (3) $\frac{\epsilon_{\rm g}\mu_{\rm g}}{r}\left(\frac{\partial u_{\rm g}}{\partial y}+\frac{\partial v_{\rm g}}{\partial r}\right)$, (4) $\frac{1}{r}\frac{\partial}{\partial\theta}\left[\epsilon_{\rm g}\mu_{\rm g}\left(\frac{1}{r}\frac{\partial v_{\rm g}}{\partial\theta}+\frac{\partial w_{\rm g}}{\partial y}\right)\right]$ and (5) $-\frac{\partial}{\partial y}\left[\frac{2}{3}\epsilon_{\rm g}\mu_{\rm g}\left(\frac{1}{r}\frac{\partial w_{\rm g}}{\partial\theta}+\frac{u_{\rm g}}{r}\right)\right]$. Overall, terms (1) $\frac{\epsilon_{\rm g}\rho_{\rm g}u_{\rm g}v_{\rm g}}{r}$ and (2) $\frac{1}{r}\frac{\partial}{\partial\theta}\left(\epsilon_{\rm g}\rho_{\rm g}v_{\rm g}w_{\rm g}\right)$ have the largest values, which are at least one order of magnitude larger than the other three terms, irrespective of the superficial gas velocity. Thus terms (3), (4) and (5) are neglected in the future discussion.

Evaluating each term in the 2D Cartesian Eqn. (7.4) revealed that the advective terms $\nabla \cdot \left(\epsilon_{\rm g} \rho_{\rm g} \vec{V_{\rm g}} \vec{V_{\rm g}}\right)$, the viscous terms $\nabla \cdot \overline{\tau}_{\rm g}$ and the body force $\epsilon_{\rm g} \rho_{\rm g} g$ were small compared to the time derivative $\frac{\partial}{\partial t} \left(\epsilon_{\rm g} \rho_{\rm g} v_{\rm g}\right)$, pressure gradient $-\frac{\partial P_{\rm g}}{\partial y}$ and drag force $F_{\rm gs} \left(v_{\rm s} - v_{\rm g}\right)$. The same



Figure 7.21 Planes used to calculate the terms in the gas momentum equation for a 3D cylindrical coordinate system.



Figure 7.22 The contour plots of terms (1) $\frac{\epsilon_{g}\rho_{g}u_{g}v_{g}}{r}$, (2) $\frac{1}{r}\frac{\partial}{\partial\theta}(\epsilon_{g}\rho_{g}v_{g}w_{g})$, (3) $\frac{\epsilon_{g}\mu_{g}}{r}\left(\frac{\partial u_{g}}{\partial y}+\frac{\partial v_{g}}{\partial r}\right)$, (4) $\frac{1}{r}\frac{\partial}{\partial\theta}\left[\epsilon_{g}\mu_{g}\left(\frac{1}{r}\frac{\partial v_{g}}{\partial\theta}+\frac{\partial w_{g}}{\partial y}\right)\right]$ and (5) $\frac{\partial}{\partial y}\left[\frac{2}{3}\epsilon_{g}\mu_{g}\left(\frac{1}{r}\frac{\partial w_{g}}{\partial\theta}+\frac{u_{g}}{r}\right)\right]$ for a superficial gas velocity 1.25U_{mf} averaged over 10–20 seconds with 3D cylindrical simulations.



Figure 7.23 The contour plots of terms (1) $\frac{\epsilon_{g}\rho_{g}u_{g}v_{g}}{r}$, (2) $\frac{1}{r}\frac{\partial}{\partial\theta}(\epsilon_{g}\rho_{g}v_{g}w_{g})$, (3) $\frac{\epsilon_{g}\mu_{g}}{r}\left(\frac{\partial u_{g}}{\partial y}+\frac{\partial v_{g}}{\partial r}\right)$, (4) $\frac{1}{r}\frac{\partial}{\partial\theta}\left[\epsilon_{g}\mu_{g}\left(\frac{1}{r}\frac{\partial v_{g}}{\partial\theta}+\frac{\partial w_{g}}{\partial y}\right)\right]$ and (5) $\frac{\partial}{\partial y}\left[\frac{2}{3}\epsilon_{g}\mu_{g}\left(\frac{1}{r}\frac{\partial w_{g}}{\partial\theta}+\frac{u_{g}}{r}\right)\right]$ for a superficial gas velocity 1.86 U_{mf} averaged over 10–20 seconds with 3D cylindrical simulations.



Figure 7.24 The contour plots of terms (1) $\frac{\epsilon_{g}\rho_{g}u_{g}v_{g}}{r}$, (2) $\frac{1}{r}\frac{\partial}{\partial\theta}(\epsilon_{g}\rho_{g}v_{g}w_{g})$, (3) $\frac{\epsilon_{g}\mu_{g}}{r}\left(\frac{\partial u_{g}}{\partial y}+\frac{\partial v_{g}}{\partial r}\right)$, (4) $\frac{1}{r}\frac{\partial}{\partial\theta}\left[\epsilon_{g}\mu_{g}\left(\frac{1}{r}\frac{\partial v_{g}}{\partial\theta}+\frac{\partial w_{g}}{\partial y}\right)\right]$ and (5) $\frac{\partial}{\partial y}\left[\frac{2}{3}\epsilon_{g}\mu_{g}\left(\frac{1}{r}\frac{\partial w_{g}}{\partial\theta}+\frac{u_{g}}{r}\right)\right]$ for a superficial gas velocity 4.04 U_{mf} averaged over 10–20 seconds with 3D cylindrical simulations.

conclusion was observed for the corresponding cylindrical terms in Eqn. (7.5). Based on this conclusion, these particular terms are not considered an important influence on the gas-solid dynamics.

Figures 7.25, 7.26 and 7.27 show the contour plots of terms (6) $-\frac{\partial P_g}{\partial y}$, (7) $\frac{\partial}{\partial t} (\epsilon_g \rho_g v_g)$ and (8) $F_{\rm gs} (v_{\rm s} - v_{\rm g})$ averaged over 10–20 seconds for three different superficial gas velocities with the 3D cylindrical simulations. The same terms for the 2D Cartesian coordinate system are plotted in Figs. 7.28, 7.29 and 7.30. The pressure gradient (6) $-\frac{\partial P_g}{\partial y}$ is similar irrespective of the superficial gas velocity and coordinate systems used. Terms (7) $\frac{\partial}{\partial t} (\epsilon_g \rho_g v_g)$ and (8) $F_{\rm gs} (v_{\rm s} - v_{\rm g})$ increase as the superficial gas velocity increases from $1.25U_{\rm mf}$ to $4.04U_{\rm mf}$ for both 3D cylindrical and 2D Cartesian simulations. For example, term (7) $\frac{\partial}{\partial t} (\epsilon_g \rho_g v_g)$ is one order magnitude smaller than (5) $-\frac{\partial P_g}{\partial y}$ with a superficial gas velocity $1.25U_{\rm mf}$. It has the same magnitude as (5) $-\frac{\partial P_g}{\partial y}$ for $4.04U_{\rm mf}$. The terms (6) $\frac{\partial}{\partial t} (\epsilon_g \rho_g v_g)$ and (7) $F_{\rm gs} (v_{\rm s} - v_{\rm g})$ have larger values in the 3D cylindrical simulations than the 2D Cartesian simulations for the same superficial gas velocity.

The difference of variable $v_{\rm g}$ calculated from the Eqs. (7.4) and (7.5) depends on comparison of the important additional terms, (1) $\frac{\epsilon_{\rm g}\rho_{\rm g}u_{\rm g}v_{\rm g}}{r}$ and (2) $\frac{1}{r}\frac{\partial}{\partial\theta}(\epsilon_{\rm g}\rho_{\rm g}v_{\rm g}w_{\rm g})$, and large common terms, (6) $-\frac{\partial P_{\rm g}}{\partial y}$, (7) $\frac{\partial}{\partial t}(\epsilon_{\rm g}\rho_{\rm g}v_{\rm g})$ and (8) $F_{\rm gs}(v_{\rm s}-v_{\rm g})$. The ratio of the additional terms to the common terms increases with the superficial gas velocity. Thus the difference of $v_{\rm g}$ calculated using the 3D cylindrical and 2D Cartesian simulations becomes larger due to increasing values of the additional terms, which was shown in §7.2.3. For the superficial gas velocity $1.25U_{\rm mf}$, the values of the additional terms are small compared with the common terms where agreement of $v_{\rm g}$ was observed for the 2D and 3D simulations. However, discrepancy was obtained with $4.04U_{\rm mf}$ due to higher values of the additional terms.



Figure 7.25 The contour plots of terms (6) $-\frac{\partial P_{\rm g}}{\partial y}$, (7) $\frac{\partial}{\partial t} (\epsilon_{\rm g} \rho_{\rm g} v_{\rm g})$ and (8) $F_{\rm gs} (v_{\rm s} - v_{\rm g})$ for a superficial gas velocity 1.25 $U_{\rm mf}$ averaged over 10–20 seconds with 3D cylindrical simulations.



Figure 7.26 The contour plots of terms (6) $-\frac{\partial P_{\rm g}}{\partial y}$, (7) $\frac{\partial}{\partial t} (\epsilon_{\rm g} \rho_{\rm g} v_{\rm g})$ and (8) $F_{\rm gs} (v_{\rm s} - v_{\rm g})$ for a superficial gas velocity 1.86 $U_{\rm mf}$ averaged over 10–20 seconds with 3D cylindrical simulations.



Figure 7.27 The contour plots of terms (6) $-\frac{\partial P_{\rm g}}{\partial y}$, (7) $\frac{\partial}{\partial t} (\epsilon_{\rm g} \rho_{\rm g} v_{\rm g})$ and (8) $F_{\rm gs} (v_{\rm s} - v_{\rm g})$ for a superficial gas velocity 4.04 $U_{\rm mf}$ averaged over 10–20 seconds with 3D cylindrical simulations.



Figure 7.28 The contour plots of terms (6) $-\frac{\partial P_{\rm g}}{\partial y}$, (7) $\frac{\partial}{\partial t} (\epsilon_{\rm g} \rho_{\rm g} v_{\rm g})$ and (8) $F_{\rm gs} (v_{\rm s} - v_{\rm g})$ for a superficial gas velocity 1.25 $U_{\rm mf}$ averaged over 10–20 seconds with 2D Cartesian simulations.



Figure 7.29 The contour plots of terms (6) $-\frac{\partial P_{\rm g}}{\partial y}$, (7) $\frac{\partial}{\partial t} (\epsilon_{\rm g} \rho_{\rm g} v_{\rm g})$ and (8) $F_{\rm gs} (v_{\rm s} - v_{\rm g})$ for a superficial gas velocity 1.86 $U_{\rm mf}$ averaged over 10–20 seconds with 2D Cartesian simulations.



Figure 7.30 The contour plots of terms (6) $-\frac{\partial P_{\rm g}}{\partial y}$, (7) $\frac{\partial}{\partial t} (\epsilon_{\rm g} \rho_{\rm g} v_{\rm g})$ and (8) $F_{\rm gs} (v_{\rm s} - v_{\rm g})$ for a superficial gas velocity 4.04 $U_{\rm mf}$ averaged over 10–20 seconds with 2D Cartesian simulations.

7.3.2 2D Cartesian and 3D Cartesian Coordinates

For the 3D Cartesian coordinate system, Eqn. (3.3) becomes:

$$\frac{\partial}{\partial t} (\epsilon_{g} \rho_{g} v_{g}) + \frac{\partial}{\partial x} (\epsilon_{g} \rho_{g} u_{g} v_{g}) + \frac{\partial}{\partial y} (\epsilon_{g} \rho_{g} v_{g} v_{g}) + \frac{\partial}{\partial z} (\epsilon_{g} \rho_{g} v_{g} w_{g}) = -\frac{\partial P_{g}}{\partial y}$$
(7.6)
$$+ \frac{\partial}{\partial x} \left[\epsilon_{g} \mu_{g} \left(\frac{\partial u_{g}}{\partial y} + \frac{\partial v_{g}}{\partial x} \right) \right] + \frac{\partial}{\partial y} \left(2 \epsilon_{g} \mu_{g} \frac{\partial v_{g}}{\partial y} \right) + \frac{\partial}{\partial z} \left[\epsilon_{g} \mu_{g} \left(\frac{\partial v_{g}}{\partial z} + \frac{\partial w_{g}}{\partial y} \right) \right]$$
$$- \frac{\partial}{\partial y} \left[\frac{2}{3} \epsilon_{g} \mu_{g} \left(\frac{\partial u_{g}}{\partial x} + \frac{\partial v_{g}}{\partial y} + \frac{\partial w_{g}}{\partial z} \right) \right] + F_{gs} (v_{s} - v_{g}) - \epsilon_{g} \rho_{g} g$$

Compared to Eqn. (7.4), one additional term appears on the left hand side of Eqn. (7.6), $\frac{\partial}{\partial z} (\epsilon_{g} \rho_{g} v_{g} w_{g})$. Two additional terms on the right hand side of Eqn. (7.6) are $\frac{\partial}{\partial z} \left[\epsilon_{g} \mu_{g} \left(\frac{\partial v_{g}}{\partial z} + \frac{\partial w_{g}}{\partial y} \right) \right]$ and $\frac{\partial}{\partial y} \left(\frac{2}{3} \epsilon_{g} \mu_{g} \frac{\partial w_{g}}{\partial z} \right)$. The contour plots of three velocities are shown in Fig. 7.31. Figure 7.32 shows the planes used to calculate the terms for 3D Cartesian coordinate system, where k is the central plane in the computational domain. By calculation, the term $\frac{\partial}{\partial z} \left(\epsilon_{g} \rho_{g} v_{g} w_{g} \right)$ has the largest value among those additional terms than the 2D Cartesian calculations and is shown in Fig. 7.33. For the common terms plotted in Fig. 7.34, (6) $-\frac{\partial P_{g}}{\partial y}$ has same values as those obtained from 3D cylindrical (Fig. 7.25) and 2D Cartesian (Fig. 7.28) coordinate systems. The values of terms (6) $-\frac{\partial P_{g}}{\partial y}$, (7) $\frac{\partial}{\partial t} (\epsilon_{g} \rho_{g} v_{g})$ and (8) $F_{gs} (v_{s} - v_{g})$ are shown in Fig. 7.34. The additional term $\frac{\partial}{\partial z} (\epsilon_{g} \rho_{g} v_{g} w_{g})$ is much smaller than the three common terms in Fig. 7.34 and can be neglected. Comparing the terms using 2D Cartesian simulations in Fig. 7.28 with the terms using 3D Cartesian simulations in Fig. 7.34, the terms have the similar values.

7.3.3 Discussion

The results in §7.2.3 show satisfactory qualitative agreement for gas velocities in the streamwise direction for the 2D and 3D simulations for both rectangular and cylindrical fluidized beds for bubbling fluidization. However, a larger discrepancy is obtained with higher superficial gas velocities in the slugging and turbulent regimes. For the slugging fluidized bed, the vertical gas velocities are underpredicted when using the 2D Cartesian system as compared to the 3D cylindrical system. A lower gas velocity is predicted with the 2D Cartesian simulations than the 3D cylindrical simulations for the turbulent regime. The lower predicted velocities for these



Figure 7.31 The contour plots of gas velocities in (a) r- (b) y- and (c) $\theta-$ directions for a superficial gas velocity $1.25U_{\rm mf}$ averaged over 10–20 seconds with 3D Cartesian simulations.

two flow regimes can be attributed to the additional terms modeled in a 3D cylindrical domain. For example, the additional terms, (1) and (2), increase the axial gas velocity. Therefore, it is not surprising that for higher superficial gas velocities, the 2D Cartesian coordinate does not accurately predict the hydrodynamics.

For the bubbling regime (superficial gas velocity $1.25U_{\rm mf}$), the pressure gradient has the largest value among all common terms. The value of pressure gradient is same for the 2D Cartesian, 3D cylindrical and 3D Cartesian coordinate systems shown in Figs. 7.25, 7.28 and 7.34. The values of additional terms appearing in the 3D cylindrical coordinate (Fig. 7.22) and the 3D Cartesian coordinate (Fig. 7.33) systems are small compared to the pressure gradient (6) $-\frac{\partial P_g}{\partial y}$. Therefore, variables $v_{\rm g}$ calculated from the equations for three different coordinate systems is dominated by the pressure gradient. The values of additional terms are negligible. Good agreement is observed between 2D and 3D simulations.

However, the magnitude of additional terms increases as the superficial gas velocity increases, refer to Figs. 7.22, 7.23 and 7.24. With higher gas velocity, the influence of those additional terms can not be neglected in simulations unlike with low gas velocity. The additional terms cause the values of $v_{\rm g}$ calculated with 3D cylindrical simulations to be higher



Figure 7.32 Planes used to calculate the terms in equations for 3D Cartesian coordinate system.

compared to 2D Cartesian simulations. Also, the difference between 2D and 3D simulations increase with higher gas velocities since the values of the additional terms increases. In general, the initial energy entering fluidized beds is mainly directed to defy the gravity and thus primarily axial direction (y-direction). The influence of non-axial terms can be neglected, therefore, the difference between 2D and 3D simulations is small (bubbling regimes). However, the flows become unstable and the energy diverts to non-axial terms with increasing energy. The non-axial terms become more important with higher inlet gas velocity, which causes the discrepancy increases from slugging to turbulent regimes.



Figure 7.33 The contour plots of term $\frac{\partial}{\partial z} (\epsilon_{\rm g} \rho_{\rm g} v_{\rm g} w_{\rm g})$ for a superficial gas velocity $1.25 U_{\rm mf}$ averaged over 10–20 seconds with 3D Cartesian simulations.



Figure 7.34 The contour plots of terms (6) $-\frac{\partial P_{\rm g}}{\partial y}$, (7) $\frac{\partial}{\partial t} (\epsilon_{\rm g} \rho_{\rm g} v_{\rm g})$ and (8) $F_{\rm gs} (v_{\rm s} - v_{\rm g})$ for a superficial gas velocity $1.25U_{\rm mf}$ averaged over 10–20 seconds with 3D Cartesian simulations.

CHAPTER 8. SIMULATIONS OF ELUTRIATION PHENOMENON IN FLUIDIZED BEDS

The carbon material that is entrained out of a gasifier reduces the reactor efficiency for biomass fluidized bed reactors. It is important to be able to predict and characterize elutriation of solid materials in order to design gas cleaning equipment, such as cyclones. In this chapter, the experimental findings for elutriation phenomena in fluidized beds are introduced first. Then, as the first step to improve the efficiency of a biomass gasifier, the capability of MFIX to simulate elutriation phenomenon in fluidized beds is validated. Quantitative data, such as elutriation rate, are compared with the experiments performed by Leva (1951) and those performed at Iowa State University (ISU).

8.1 Background

Biomass is a term for all organic materials that stems from plants (Williams et al., 2001). Biomass is produced by green plants combining CO_2 in the air, water and sunlight via photosynthesis to generate carbohydrates. The solar energy is stored in the chemical bonds of a biomass material. Recently, biomass has gained consideration as a renewable energy source due to several reasons (McKendry, 2002a). First, global warming is believed to cause climatic changes due to high emission levels of greenhouse gases in which CO_2 is the most important concern. When energy stored in the chemical bonds of biomass is extracted, carbon is oxidized to produce CO_2 and water. Roughly the same amount of carbon emitted during conversion is taken up during plant growth. The use of biomass therefore does not contribute to the build up of CO_2 in the atmosphere. However, it takes millions of years to generate fossil fuels such as coal and oil. Thus, fossil fuels are not renewable within a time-scale that mankind can use since burning fossil fuels uses "old" biomass and converts it into "new" CO_2 , which contributes to the greenhouse effect. Second, technological developments have increased the yields of biomass and made the conversion of energy from biomass more efficient than before. For example, the world crop yield of sorghum, corn, rye, rice and wheat increased by 20.1%, 19.5%, 30.1%, 8.7% and 4.5%, respectively, during the period 1991–1996 (Sadaka et al., 2002). The cost of electricity produced from low cost biomass residues and high conversion efficiency can be competitive with fossil fuel-based power generation. Other reasons that biomass is becoming a more viable energy source are the increasing price of fossil fuels and food surpluses in Western Europe and the US.

Biomass can be converted into three main types of product: power/heat generation, transport fuel and chemical feedstock. The first two products are related to energy which can be generated by three technologies: thermo-chemical, bio-chemical/biological and mechanical extraction. Within thermo-chemical conversion four processes are available: combustion, pyrolysis, gasification and liquefaction/hydro-thermal upgrading. Among those processes, gasification by fluidized beds is the focus point in this study. Fluidized beds have been used extensively for coal gasification for many years. Fluidized beds provide a promising technology to convert biomass to energy as a result of well-mixed solids and nearly isothermal conditions (McKendry, 2002b). Gasification converts biomass into a combustible gas mixture by the partial oxidation of biomass at a high temperature, usually around 800–900 °C. Gasification can be classified depending on the gasifying agent such as air, steam, steam-oxygen, air-steam, oxygen-enriched air, etc (Lv et al., 2004). The gas produced can be used to generate the methanol and hydrogen or burned in equipments such as gas turbines. The biomass integrated gasification and combined cycle (BIGCC) is one promising concept since gas turbines convert the gaseous fuel to electricity with a high overall conversion efficiency.

The methodology that is available for coal is often applied to biomass combustion and gasification (Williams et al., 2001) although there are still areas where there is a lack of information. Biomass reactions follow four fundamental steps: heating-up of biomass particles, devolatilization, reactions of volatiles and char reactions. However, biomass has a great dif-

Biomass	Moisture	Volatile	Fixed	Ash
		matter	carbon	
Wood	20	82	17	1
Wheat straw	16	59	21	4
Barley straw	30	46	18	6
Bituminous coal	11	35	45	9

Table 8.1Proximate analysis of some biomass and coal (wt %) (McKendry,
2002a).

ference in chemical and physical properties from that of fossil fuels (Scala and Salatino, 2002; Scala and Chirone, 2004). Biomass usually has a higher moisture and volatile content, a more porous and fragile structure, a lower density and a higher intrinsic reactivity. The proximate analysis of some biomass and coal is shown in Tab. 8.1. The loosely connected structures of chars after devolatilization form carbon fines by comminution such as attrition and fragmentation. Attrition and fragmentation are far more extensive in the case of high-volatile fuels. The reactor efficiency is directly determined by the relative extent of the combustion time scale and of the residence time of carbon fines produced by comminution and reactions in the bed.

8.2 Introduction to Elutriation in Fluidized Beds

The entrainment of carbon materials from the freeboard is the main source of loss in reactor efficiency for fluidized bed gasifiers or combustors, especially for biomass reactors. In addition, the fines exiting from fluidized beds due to a size distribution of input solids or by attrition must either be recovered or removed from the gas for economic or environmental requirements. The elutriation of solid materials has to be known to design gas cleaning equipment.

The fluidized bed may be divided into different zones as shown in Fig 8.1. Above the distributor is the dense fluidized bed, where bubbles grow and rise to the surface of the bed. When bubbles burst at the surface, solids are thrown above the bed and entrained by the gas in the freeboard. The freeboard height is defined as the height between the surface of the dense bed and the gas outlet. The height of the column is usually prescribed to prevent the



Figure 8.1 Different zones in a fluidized bed.

solids from being carried out of the vessel. The entrainment rate gradually decreases until it becomes constant or nearly constant from experimental observations. The height where the entrainment rate becomes constant is called the transport disengaging height (TDH).

8.2.1 Elutriation Rate

The rate of elutriation of fines from fluidized beds can be expressed as a first order equation (Leva, 1951):

$$\begin{pmatrix} \text{rate of removal} \\ \text{of solids of size } d_i \end{pmatrix} = K \begin{pmatrix} \text{weight of that size} \\ \text{solids in the bed} \end{pmatrix}$$

and

$$-\frac{\mathrm{d}W_i}{\mathrm{d}t} = KW_i \tag{8.1}$$

where W_i is the weight of solids of size d_i in the bed. Earlier researchers (e.g. Osberg and Charlesworth (1951)) correlated the value of K (sec⁻¹), the elutriation constant, with particle diameter, superficial gas velocity and so on.

The rate of elutriation (Yagi and Aochi, 1955) also can be defined as:

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$$\begin{pmatrix} \text{rate of removal of solids} \\ \text{of size } d_i \text{ per area of bed} \\ \text{surface} \end{pmatrix} = K^* \begin{pmatrix} \text{fraction of bed} \\ \text{consisting of} \\ \text{size } d_i \end{pmatrix}$$

That is:

$$-\frac{1}{A}\frac{\mathrm{d}W_i}{\mathrm{d}t} = K^*\left(\frac{W_i}{W}\right) \tag{8.2}$$

where A is the bed cross section and W is the weight of all solids in the bed. K^* (kg/m² · sec) is called the elutriation rate constant, which is independent of bed height. Comparing Eqs. (8.1) and (8.2),

$$K^* = \frac{KW}{A} \tag{8.3}$$

Most researchers, for example Wen and Hashinger (1960), Tanaka et al. (1972) and Baeyens et al. (1992), related K^* with system parameters to describe the elutriation.

Both batch systems and steady state flow systems have been used to study the elutriation phenomena (Kunii and Levenspiel, 1991). For a batch system with two components such as in this study, Eqs. (8.1) and (8.2) can be expressed in terms of the concentration of fines $X_{\rm F}$:

$$-\frac{\mathrm{d}X_{\mathrm{F}}}{\mathrm{d}t} = \frac{K^*A}{W}X_{\mathrm{F}} = KX_{\mathrm{F}}$$
(8.4)

The bed weight W can be considered constant if the fraction or concentration of fines is small, usually less than 20%. Thus integration of Eqn. (8.4) gives:

$$X_{\rm F} = X_{\rm F0} \, \exp\left(-\frac{K^*A}{W}t\right) = X_{\rm F0} \, \exp\left(-Kt\right) \tag{8.5}$$

where $X_{\rm F0}$ is the initial fine concentration in the bed. Experiments have demonstrated that K^* is close to a constant for binary mixtures with a small concentration of fines in batch systems. Thus according to Eqn. (8.5), the slope of the data on a semi-log plot of $X_{\rm F}/X_{\rm F0}$ versus time gives the elutriation rate K^* . From experimental observation, the straight line in the semi-log plot usually anchors at one point named the critical concentration $X_{\rm F}^*$. Leva and Wen (1971) stated that the transport of fines through the bed to the upper surface became rate controlling after $X_{\rm F}$ reached the critical concentration. Only the initial straight portion of the data is used to calculate elutriation rate constants.

8.2.2 Entrainment at the Bed Surface

The particles are ejected from the fluidized bed surface into the freeboard primarily due to the eruption of bubbles. A dominant mechanism by which particles elutriate is not clearly established (Wen and Chen, 1982). Two possible explanations which are reported to be responsible for the particle ejection are (Yang, 2003):

- 1. Solids held in the leading roof or nose of a bubble are projected into the freeboard after the bubble breaks.
- 2. The particles transported in the bubble wake are thrown upwards when the bubbles burst at the surface.

Some researchers (Zenz and Weil, 1958; Do et al., 1972) concluded that the nose mechanism is the most important. The wake mechanism was considered to be the dominating mechanism by Yates and Rowe (1977) and George and Grace (1978). Pemberton and Davidson (1986) stated that both mechanisms may be used to explain that ejection of particles into the freeboard. For example, the nose mechanism prevails when the Geldart group A particles fluidized at high gas velocities, whereas the wake mechanism governs the ejection of the Geldart group B particles which fluidize at low gas velocities. Also, differences exist for the solids ejected into the freeboard by the two mechanisms (Yang, 2003). The solids thrown into the freeboard from the nose of bubbles are much finer than the bed materials. However, the solids thrown into freeboard from the wake of bubbles are similar to the size distribution of the bed materials.

The entrainment at the bed surface, E_0 , can be obtained by extrapolation of the entrainment rate data in the freeboard to the bed surface (Wen and Chen, 1982). Table 8.2 lists some of the correlations in the literature for the entrainment rate at the bed surface, where d_b is the bubble diameter at the bed surface. The column diameter is used instead of bubble diameter for slugging beds. The subscript mf indicates properties at minimum fluidization.

Equation	Reference
$E_0 = 3d_{\rm p} \frac{(1 - \epsilon_{\rm mf})(U - U_{\rm mf})}{d_{\rm b}}$ bubble nose model	Pemberton and Davidson (1986)
$E_0 = 0.1 \rho_{\rm s} (1 - \epsilon_{\rm mf}) (U - U_{\rm mf})$ bubble wake model	
$E_0 = 3.07 \times 10^{-9} \frac{Ad_{\rm b}(U - U_{\rm mf})^{2.5} \rho_{\rm g}^{3.5} g^{0.5}}{\mu^{2.5}}$	Wen and Chen (1982)
$E_0 = 9.6A(U - U_{\rm mf})^{2.5}Ad_{\rm b} \left(\frac{298}{T}\right)^{3.5}$	Choi et al. (1989)

Table 8.2 Correlations for the entrainment at the bed surface (Yang, 2003).

8.2.3 Elutriation of Fines Above TDH

As mentioned earlier, the entrainment flux becomes almost constant above the transport disengaging height (TDH). The entrainment and elutriation rate constants above TDH usually have the index " ∞ ", such as E_{∞} and K_{∞}^* . The effect of various parameters on the elutriation of fines above TDH will be presented in the following paragraphs.

• Effect of Bed Hydrodynamics. Since the entrainment rate at the bed surface is associated with bubble size and frequency in the bed, the bed hydrodynamics has a more pronounced effect on the entrainment rate at the bed surface. However, the elutriation rate constant above TDH is not influenced by the bed dynamics. Sanari and Kunii (1962) stated that no strong influence on the elutriation of fines was found with different coarse particle sizes (from 300μ m to 2500μ m). Tanaka et al. (1971) reported that no effect was found with two different distributor plates. The elutriation rates were nearly equal with both totally mixed and separated systems (Lin et al., 1980). Choi et al. (1989) concluded that the bed height had little effect on the elutriation rate. Thus, Wen and Chen (1982) drew conclusions that the elutriation rate above TDH is not affected strongly by the hydrodynamics of bed.

- Effect of Gas Velocity. The influence of the superficial velocity on the elutriation rate constant is tremendous. The velocity has an effect on both entrainment rate on the bed surface and elutriation rate. In general, the elutriation rate constant is proportional to the 2nd to 6th power of the gas velocity. At the higher velocities, the power reduces to less than one (Wen and Chen, 1982).
- Effect of Fine Particle Size. Most researchers agree that the elutriation rate constant increases with decreasing fine particle size. However, a trend is observed that the elutriation rate constant levels off or decreases with decreasing size for group A particles, which is probably due to the formation of cohesive fine particles.
- Effect of Column Diameter. There is a lack of data available on the influence of scale up. Lewis et al. (1962) tested elutriation with fluidized bed diameters ranging from 0.019 to 0.146 m. They concluded that the elutriation rate became independent of bed size for a diameter larger than 0.1 m. However, the elutriation rate increased with decreasing diameter for a smaller bed. Others, such as Colakyan and Levenspiel (1984) and Tasirin and Geldart (1998), confirmed the conclusions of Lewis et al. (1962). The reason is that fine particles tend to fall back into the bed along the wall. Thus less fine particles descend if the column diameter is large, which causes higher elutriation rate for larger beds.
- Effect of Temperature and Pressure. Chan and Knowlton (1984) investigated the influence of pressure. They found that the elutriation increases with pressure due to the increasing gas density. For the effect of temperature, George and Grace (1981) did not find an influence of temperature on entrainment. Choi et al. (1997) stated that there was a minimum entrainment for temperatures from 450 to 700 K. However, Wouters and Geldart (1998) did not find a minimum for the entrainment rate, and concluded that the elutriation rate constant decreased with increasing temperature.

Table 8.3 lists some empirical correlations available in the literature for the elutriation rate constant above TDH. The parameters U and u_t are the superficial gas velocity and the terminal velocity of fines, respectively, d_p is the fine diameter and D_h is the hydraulic diameter of the fluidized bed column. *Re* and *Re*_t are the Reynolds numbers based on bed column and fine particles, respectively. Yang (2003) plotted the elutriation rate constant versus gas velocity using different correlations. A significant difference in magnitude and predicted tendencies is observed. Wen and Chen (1982) concluded that the deviation of some of the experimental data was greater than $\pm 50\%$ due to the difficulty in obtaining accurate elutriation data. Therefore, the selection of correlations should be careful due to the diversity of the experimental conditions.

8.2.4 Entrainment Below TDH

An exponential decay of the entrainment rate with respect to freeboard height h was observed by Andrews (1960). The entrainment rate of solids can be represented as:

$$E_h = E_{\infty} + (E_0 - E_{\infty}) \exp(-ah)$$
 (8.6)

where E_h is the entrainment rate of fine particles in the freeboard at a height h. It can be calculated using the elutriation rate E_{∞} , the entrainment rate at the bed surface E_0 and the decay constant a. Wen and Chen (1982) concluded that a varies between 2.2 to 6.6 m⁻¹. The value of 4.0 m⁻¹ is recommended for a system in which no information is available.

8.2.5 Estimation of TDH

The freeboard height should be higher than the TDH in order to reduce the elutriation of fines. However, increasing the freeboard height excessively beyond the TDH has no advantage. Currently there is no commonly accepted calculation for estimating the TDH, however, there are a number of correlations that exist in the literature and a few of them are summarized in Tab. 8.4.

Equation	Reference
$\frac{K_{\infty}^* g d_{\rm p}^2}{\mu \left(U - u_{\rm t}\right)^2} = 0.0015 R e_{\rm t}^{0.6} + 0.01 R e_{\rm t}^{1.2}$	Yagi and Aochi (1955)
$\frac{K_{\infty}^{*}}{\rho_{\rm g}U} = \begin{cases} 1.26 \times 10^{7} \left(\frac{U^{2}}{gd_{\rm p}\rho_{\rm s}^{2}}\right)^{1.88} & \text{for } \frac{U^{2}}{gd_{\rm p}\rho_{\rm s}^{2}} < 3.1\\ 1.31 \times 10^{4} \left(\frac{U^{2}}{gd_{\rm p}\rho_{\rm s}^{2}}\right)^{1.18} & \text{for } \frac{U^{2}}{gd_{\rm p}\rho_{\rm s}^{2}} > 3.1 \end{cases}$	Zenz and Weil (1958)
$\frac{K_{\infty}^{*}}{\rho_{\rm g} \left(U - u_{\rm t}\right)} = 1.52 \times 10^{-5} \left[\frac{\left(U - u_{\rm t}\right)^2}{gd_{\rm p}}\right]^{0.5} Re^{0.725} \left(\frac{\rho_{\rm s} - \rho_{\rm g}}{\rho_{\rm g}}\right)^{1.15}$	Wen and Hashinger (1960)
$\frac{K_{\infty}^{*}}{\rho_{\rm g} \left(U - u_{\rm t}\right)} = 4.6 \times 10^{-2} \left[\frac{\left(U - u_{\rm t}\right)^2}{gd_{\rm p}}\right]^{0.5} Re_{\rm t}^{0.3} \left(\frac{\rho_{\rm s} - \rho_{\rm g}}{\rho_{\rm g}}\right)^{0.15}$	Tanaka et al. (1972)
$\frac{K_{\infty}^{*}}{\rho_{\rm g}U} = 23.7 \mathrm{exp}\left(-5.4\frac{u_{\rm t}}{U}\right)$	Geldart et al. (1979)
$\frac{K_{\infty}^{*}}{\rho_{\rm g}U} = 9.43 \times 10^{-4} \left(\frac{U^2}{gd_{\rm p}}\right)^{1.65}$	Lin et al. (1980)
$K_{\infty}^{*} = 0.011 \rho_{\rm s} \left(1 - \frac{u_{\rm t}}{U} \right)^2$	Colakyan and Levenspiel (1984)
$K_{\infty}^{*} = 2.8 \times 10^{-2} \left(\frac{U - u_{\rm t}}{U}\right)^{1.6} \left(\frac{\rho_{\rm s} - \rho_{\rm g}}{\rho_{\rm s}}\right)^{0.54U_{\rm t}^{2.1}} D_{\rm h}$	Kato et al. (1985)
$\frac{K_{\infty}^{*}}{\rho_{\rm g}U} = 1.6 \left(\frac{U}{u_{\rm t}}\right) \left(1 - \frac{u_{\rm t}}{U}\right)$	Sciazko et al. (1991)
$K_{\infty}^{*} = 5.4 \times 10^{-5} \rho_{\rm s} \left(\frac{U}{0.2}\right)^{3.4} \left(1 - \frac{u_{\rm t}}{U}\right)^{2}$	Baeyens et al. (1992)
$K_{\infty}^{*} = \begin{cases} 23.7\rho_{\rm g}U^{2.5} \exp\left(-5.4\frac{u_{\rm t}}{U}\right) & \text{for } Re < 3000\\ 14.5\rho_{\rm g}U^{2.5} \exp\left(-5.4\frac{u_{\rm t}}{U}\right) & \text{for } Re > 3000 \end{cases}$	Tasirin and Geldart (1998)

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Table 8.3 Correlations for the elutriation rate constant K_{∞}^{*} (Yang, 2003).

Table 8.4 Correlations for the estimation of the transport disengaging height (TDH) (Yang, 2003).

Equation	Reference
$\text{TDH} = 0.85 U^{1.2} \left(7.33 - 1.2 \log U \right)$	Chan and Knowlton (1984)
$\text{TDH} = 1000 \frac{U^2}{g}$	Fournol et al. (1973)
$\text{TDH} = 18.2d_{\text{b}}$	George and Grace (1978)
$\text{TDH} = 13.8d_{\text{b}}$	Fung and Hamdullahpur (1993)

8.3 Benchmark Case

The first experimental work to investigate the elutriation phenomenon was performed by Leva (1951). Leva reported basic observations on the elutriation of fines from fluidized beds and proposed Eqn. (8.1) for the elutriation rate. The systems investigated were composed of one coarse and one fine component. The influence of system variables such as gas velocity, and fine and coarse particle diameters on the elutriation rate was presented. This experimental work is chosen as the benchmark case to test the feasibility of MFIX to simulate the elutriation phenomenon.

8.3.1 Simulation Conditions and Grid Resolution Study

Figure 8.2(a) represents the experimental setup (Leva, 1951). It is a cylinder with a diameter of 3.4 cm and height of 121.9 cm. A small cyclone at the top of the unit was used to collect the fines. Two narrow cuts of particles were used in the experiments. The fines were elutriated ahead of time in order to remove the finer particles. In the experiments, 80% coarse particles and 20% fines were mixed first before being charged into the fluidized bed. The initial


Figure 8.2 (a) Fluidized bed used for elutriation experiments and (b) computational domain used in simulations.

bed weight was 200 g and the static bed height was 11.3 cm. Gas with a low velocity was introduced into the bed to fluidize the solids in order to ensure a uniform mixture before a higher velocity was used to elutriate the fines. The elutriation rate of fines was obtained for conditions above the TDH.

In the simulations, the fine and coarse particles are solved as two individual phases, each described by equations for continuity and momentum (i.e., two equations per solid phase), where each phase is characterized by a unique diameter and density. Although the experiments of Leva (1951) investigated the mixture of a combination of sand and iron catalyst particles, only a mixture of sand are simulated in this study. The coarse and fine particles are modeled with different diameters but the same sand density of 2.6 g/cm³. One experimental case is chosen to determine the correct grid size using a gas velocity of 27.4 cm/s. The diameters of

Table 8.5 The grid resolution and cell sizes.

Grid	$\Delta x (cm)$	$\Delta y (cm)$
16×240	0.21	0.50
16×480	0.21	0.25
32×480	0.105	0.25
32×960	0.105	0.125

the fine and coarse particles are 51.3 μ m and 191 μ m, respectively. Since three-dimensional simulations are computationally prohibitive, a two-dimensional Cartesian coordinate system is considered for the simulations, representing a plane for the cylindrical domain with a width of 3.4 cm shown in Fig. 8.2(b). The Superbee method (§3.2.2) is used to discretize the governing equations and the blending formulations (§7.2.2) are employed for solid stress calculations. The walls are modeled using no-slip boundary conditions for both gas and solid phases. A uniform gas velocity is specified at the bottom of the bed and ambient pressure is specified at the exit. The particles are fluidized by air at ambient conditions. For the initial condition, the fluidized bed is at minimum fluidization with a void fraction of 0.42. The freeboard is considered to be occupied only by the gas.

The grid resolution study was conducted first and the number of cells used are shown in Tab. 8.5. Figure 8.3 shows the profiles of the time-averaged void fraction and volumetric fraction of fines averaged over the x-direction versus bed height. An obvious difference is observed when the grid is changed from 16×240 to 16×480 for both variables. However, there are no significant changes in the void fraction and volumetric fraction of fines for grids 16×480 , 32×480 and 32×960 . Therefore, a uniform grid of 16×480 is employed in all the simulations for the benchmark case. Figure 8.3(a) indicates that the bed expands to an approximate height of 22 cm. The exponential decay of fines in the freeboard is observed from 22 to 30 cm in 8.3(b). Highest volumetric fraction of fines appears at a bed height about 10 cm and gradually reduces as the bed height decreases, which is an indication of segregation in the dense bed. Fines float on top of the bed and coarse particles accumulate at the bottom.



U.4

Bed height (cm)



1.2



Figure 8.3 (a) Void fraction and (b) volumetric fraction of fines averaged over 10–20 s and x- direction versus bed height for different grids.

8.3.2 Elutriation Rate Constants Above TDH

As stated in §8.2.3, the elutriation rate constants above TDH vary with different parameters. The influence of gas velocity and fine and coarse particle diameters on K_{∞}^* is presented next.

8.3.2.1 Effect of Gas Velocity

Since the influence of gas velocity is dominant, its effect on the elutriation rate constant K_{∞}^* is investigated first. In the experiments (Leva, 1951) and the simulations, the diameters of fine and coarse particles are 51.3 μ m and 191 μ m, respectively. The three different gas velocities studied are 21.3 cm/s, 27.4 cm/s and 32.9 cm/s, respectively.

In the experiments, the fixed rate of gas flow was maintained for sufficient time to drive a small portion of the fines out of the system. The time for test runs in the experiments with gas velocities 21.3 cm/s and 27.4 cm/s was near 60 mins and with 32.9 cm/s was 40 mins. According to Leva and Wen (1971), the slope of the initial linear portion of the data on a semilog plot of $X_{\rm F}/X_{\rm F0}$ versus time is used to calculate elutriation rate constants. The run time used to obtain the elutriation rate constants was about 20 mins for all three cases. However, for the simulations herein, the computational time required to perform calculations up to a physical time of 20 mins for each case would be about four months using 4 processors on a DEC Alpha EV6.7 cluster running at 833 MHz, even for two-dimensional simulations. Therefore, the second task with respect to the simulations is to determine the necessary physical time to obtain a reasonable elutriation rate constant, which can accurately predict the trends for elutriation rate constants with varying system parameters. An intermediate gas velocity of 27.4 cm/s used in the experiments is selected for this task and this case is simulated up to 600 s (10 mins).

Figure 8.4 shows the plot of $\ln(X_F/X_{F0})$ versus time for gas velocity 27.4 cm/s. At the initial stage of the simulation, X_F is zero since fines do not elutriate from the bed at startup. Neglecting the initial startup (about 12 s), a linear line is observed up to 600 s and can be used to calculate the elutriation rate, K_{∞} . The slope of the line, K_{∞} , is obtained by linear



Figure 8.4 $\ln(X_{\rm F}/X_{\rm F0})$ versus time for gas velocity 27.4 cm/s.

regression using EXCEL and the elutriation rate constant, K_{∞}^* , is calculated using Eqn. (8.3). Elutriation rate constants versus the amount of time used to calculate the constants are plotted in Fig. 8.5 for gas velocity 27.4 cm/s. For example, using data up to time of 80 s yields K_{∞}^* of 0.0354. This is one data point in Fig. 8.5. A lower elutriation rate constant is observed when less time is used to calculate the constant, such as with 40 s and 60 s. The values of K_{∞}^* increase as more data are used. From Fig. 8.5, the fluctuation of elutriation rate constants is less than 1% after 200 s such that 200 s is sufficient to predict the elutriation rate constants. The error incurred using times between the values of 60 to 200 s to predict elutriation rate constants is less than 5%.

The plot of $\ln(X_{\rm F}/X_{\rm F0})$ versus time for three different gas velocities is shown in Fig. 8.6. The simulation time used to calculate the elutriation rate constants is 200 s for gas velocities 21.3 cm/s and 32.9 cm/s and 600 s for gas velocity 27.4 cm/s. As the gas velocity increases, the slopes increase and therefore the elutriation rate constants increase. The constants are shown in Tab. 8.6 and plotted in Fig. 8.7, where the predicted K_{∞}^* (triangles) is compared with the experiments of Leva (1951) (squares). The elutriation rate constants are proportional to approximately 5.6th power of the gas velocity. Although Leva (1951) did not give an estimation of experimental errors, Wen and Chen (1982) concluded that the deviation of some of the



Figure 8.5 Elutriation rate constants versus the amount of time used to calculate the constants for gas velocity 27.4 cm/s.



Figure 8.6 $\ln(X_{\rm F}/X_{\rm F0})$ versus time for different gas velocities.

	$K^*_{\infty} ~(\mathrm{kg/m^3s})$		
Gas velocity (cm/s)	Experiments	Simulations	
21.3	0.0058	0.0030	
27.4	0.0289	0.0367	
32.9	0.0652	0.1750	

Table 8.6 Comparison of K_{∞}^* from the experiments (Leva, 1951) and the simulations.

Table 8.7 The experimental conditions for correlations (Yang, 2003).

Reference	Gas velocity	Bed diameter	Coarse particle diameter
	(m/s)	(m)	(mm)
Yagi and Aochi (1955)	0.3 - 1.0	0.07 – 1.0	0.1 – 1.6
Wen and Hashinger (1960)	0.6 - 1.0	0.102	0.7
Tanaka et al. (1972)	0.9 – 2.8	0.031 - 0.067	0.7 – 1.9
Baeyens et al. (1992)	0.2 – 0.7		0.03 – 0.78
Tasirin and Geldart (1998)	0.2 - 0.8	0.076, 0.152	0.017 – 0.077
Simulations	0.21 – 0.33	0.0335	0.191

experimental data was greater than $\pm 50\%$ due to difficulty in obtaining accurate elutriation data. There existed a size distribution of fines in the experiments although a narrow range of particles was used. Also, the electrostatic forces are larger at higher velocities and causes the sand to adhere to the bed walls in the experiments, reducing the elutriation of sand particles.

The correlations available in the literature (listed in Tab. 8.3) are also shown in Fig. 8.7 along with the results of experiments and simulations. Significant differences are observed from the correlations since different fluidized systems were applied to derive the correlations. From Fig. 8.7, five correlations fit well with the results obtained from Leva's experiments and the simulations: Yagi and Aochi (1955) (1), Wen and Hashinger (1960) (3), Tanaka et al. (1972) (4), Baeyens et al. (1992) (10) and Tasirin and Geldart (1998) (11). The conditions for those experiments used to derive the correlations are shown in Tab. 8.7. No common trend is found for the experimental conditions.



Figure 8.7 Comparison of elutriation rate constants by correlations of Yagi and Aochi (1955) (1), Zenz and Weil (1958) (2), Wen and Hashinger (1960) (3), Tanaka et al. (1972) (4), Geldart et al. (1979) (5), Lin et al. (1980) (6), Colakyan and Levenspiel (1984) (7), Kato et al. (1985) (8), Sciazko et al. (1991) (9), Baeyens et al. (1992) (10), Tasirin and Geldart (1998) (11), experiments (12) and simulations (13).

8.3.2.2 Effect of Fine Particle Size

The influence of fine particle size is investigated with two fine diameters, 40.5 μ m and 67.1 μ m, along with 51.3 μ m presented in the previous section. The coarse particle size is 191 μ m for all cases. Three gas velocities used for the simulations are same as before: 21.3 cm/s, 27.4 cm/s and 32.9 cm/s. The cases with the fine particle sizes, 40.5 μ m and 67.1 μ m, are simulated up to 60 s.

Elutriation rate constants versus gas velocity for different fine particle sizes are plotted in Fig. 8.8. For the same gas velocity, the elutriation rate constant increases with decreasing fine particle size. The increasing of rate constants with larger gas velocity is again observed for a fixed fine particle size.



Figure 8.8 Elutriation rate constants versus gas velocity for different fine particle sizes.

8.3.2.3 Effect of Coarse Particle Size

Bed hydrodynamics is influenced by coarse particles fluidized in the vessel, affecting the entrainment rate at the bed surface. Experimental observation concludes that the elutriation rate constant above TDH is not influenced by different coarse particle sizes (Sanari and Kunii, 1962). In this study, three coarse particle sizes are chosen: 191 μ m, 227 μ m and 320 μ m. The fine particle size is 51.3 μ m for all tests. The simulations with 227 and 320 μ m run up to 60 s.

Elutriation rate constants versus gas velocity for different coarse particle sizes are shown in Fig. 8.9. The values of rate constants are similar for the same gas velocity with different sizes of coarse particles. In conclusion, elutriation rate constant does not strongly depend on coarse particle size.

8.3.3 Elutriation Rate Constants Below TDH

The entrainment model proposed in the literature, shown in Eqn. (8.6), is based on the assumption of an exponentially decaying entrainment rate along the freeboard. The data along the freeboard calculated from the simulations are chosen to fit the data in order to obtained the decay constant *a*. Figure 8.10 shows comparisons of entrainment rate from the simulations



Figure 8.9 Elutriation rate constants versus gas velocity for different coarse particle sizes.

with the proposed model for different gas velocities. Fine and coarse particle sizes are 51.3 μ m and 191 μ m are used for all of the simulations. Values between 2.2 m⁻¹ and 6.6 m⁻¹ were determined from the experiments (Wen and Chen, 1982). However, a constant between 0.4 m⁻¹ and 0.8 m⁻¹ is obtained from the simulations. Thus, a faster decay of fines is observed in the experiments above the bed surface. The improved constituent models for the interaction of gas-solid phases and solid-solid phases could reduce the difference between experiments and simulations.

8.4 Experiments Performed at Iowa State University (ISU)

Experimental investigation of the elutriation phenomenon in fluidized beds is performed in the fluidized bed lab at Iowa State University. Figure 8.11(a) represents the setup for the elutriation experiments. The column has a diameter of 9.5 cm and height of 86.1 cm. Both fine and coarse particles are glass beads with a density of 2.6 g/cm³. A composition of 15% fines and 85% coarse particles is mixed before being placed into the vessel. The weights of fine and coarse particles are about 175 and 1000 g, respectively. Air enters from the bottom of the bed after passing a humidifier in order to reduce electrostatic forces during fluidization,



Figure 8.10 Comparison of entrainment rate from the simulations with the proposed model for different gas velocities.

therefore, reducing the possibility of an accumulation of particles on the wall of the vessel. An expansion part at the top of the vessel is used to collect the fines by decreasing the air velocity.

The computational domain for the simulations is shown in Fig. 8.11(b). Two-dimensional simulations are performed employing the Cartesian coordinate system. The fine and coarse particles used in the experiments have a size distribution between 500–600 μ m and 180–212 μ m, respectively. In the simulations, the fine and coarse particles are solved as two individual phases where each phase is characterized by a unique diameter. Thus, the standard averaged diameters of coarse and fine particles, 537 μ m and 196 μ m, are used in simulations for two solid phases. The Superbee method is used to discretize the governing equation; and the blending formulations are employed for solid stress calculations. The walls are modeled using no–slip boundary conditions for both gas and solid phases and an ambient pressure is specified at the exit. For the initial condition, the dense bed is at the minimum fluidization with a void fraction 0.42 and the freeboard is occupied by the gas. The number of cells is 40 × 300 in x- and y- directions, respectively, which is about 5 times the coarse particle diameter. Thus, the height of the freeboard is lower than the TDH, therefore, elutriation rate constants (K^*) obtained



Figure 8.11 (a) Fluidized bed used for experiments (courtesy of Nathan Franka) and (b) computational domain used in simulations.

should be larger than those above the TDH, K_{∞}^* .

Four different gas velocities, 1.8, 2.0, 2.2 and 2.5 m/s, are used in the experiments to investigate the influence of gas velocity on elutriation rate constants. The comparison of the results for the experiments and the simulations is shown in Figs. 8.12, 8.13, 8.14 and 8.15, which corresponds to the four gas velocities. The solid lines and lines with symbols indicate the simulation and experiment results, respectively. Each symbol represents a time that the amount of elutriated fines is measured experimentally. Different time intervals used to obtain the experimental data have little effect on the results, which is observed from Figs. 8.12–8.15, indicating the repeatability of the experiments and providing confidence in the experimental procedure. All simulations run up to the same time as the experiments. The initial slope at the beginning of a simulation is larger, which is caused by the start-up of the bed and is more obvious with higher gas velocity (Figs. 8.14 and 8.15). The initial part of data is not considered when calculating elutriation rate constants. The slope, which indicates the elutriation rate constant, increases as gas velocity increases from 1.8 m/s (Fig. 8.12) to 2.5 m/s (Fig. 8.15). The elutriation rate constants obtained from the experiments and the simulations are shown



Figure 8.12 $\ln(X_{\rm F}/X_{\rm F0})$ versus time for gas velocity 1.8 m/s.

Table 8.8	Comparison of K^*	from	the	experiments	performed	at	ISU	and
	the simulations.							

	$K^* (\mathrm{kg/m^3s})$		
Gas velocity (m/s)	Experiments	Simulations	
1.8	1.982	1.550	
2.0	3.023	2.359	
2.2	5.333	3.981	
2.5	7.717	7.185	

in Tab. 8.8 and compared in Fig. 8.16. The results of the simulations show good agreement with the experimental results for all four gas velocities.



Figure 8.13 $\ln(X_{\rm F}/X_{\rm F0})$ versus time for gas velocity 2.0 m/s.



Figure 8.14 $\ln(X_{\rm F}/X_{\rm F0})$ versus time for gas velocity 2.2 m/s.



Figure 8.15 $\ln(X_{\rm F}/X_{\rm F0})$ versus time for gas velocity 2.5 m/s.



Figure 8.16 Comparison of elutriation rate constants obtained from the experiments and the simulations.

CHAPTER 9. CONCLUSIONS AND FUTURE WORK

Gas-solid fluidized bed reactors have many industrial applications due to their advantageous characteristics such as excellent gas-solid contacting and high rate of heat transfer. With increasing computational facilities, CFD can be a viable tool to assist the design of reactors. However, CFD modeling for reacting gas-solid flows is very complex and not widely applied. In this research, the effort has been put to advance the progress in modeling multiphase reacting flows using a finite-volume CFD code (MFIX), which models chemical reactions and heat transfer in fluid-solid flows. The goal of this study was to improve computations for solving complex chemistry problems and capture elutriation phenomena in fluidized beds. In addition, the capability of the code (MFIX) to predict actual physics using 2D modeling was also investigated.

The isothermal silane pyrolysis case was first used to investigate the in-situ adaptive tabulation (ISAT) performance with the modified MFIX-ISAT algorithm. With MFIX-ISAT timesplitting algorithm, ISAT which can treat complex chemistry efficiently was employed to solve the chemical reactions in fluidized bed reactors; whereas the hydrodynamics of multiphase flows was resolved by MFIX. At first, ISAT was called by MFIX at every time step during the calculation. An error tolerance (E) of 10^{-5} was chosen for ISAT to maintain the accuracy of computational results for the isothermal cases. In general, ISAT(DI) was in very good agreement with MFIX, which indicated the successful implementation of the time-splitting method of MFIX with ISAT. Although the results for DI and ISAT compared well, ISAT calculations did not achieve a good performance. A relatively large number of records were stored in the table and the table did not reach maturity at the end of the simulation. The poor performance of ISAT was due to the drastically varying time step in MFIX, since ISAT determines the EOA based both on the distance in the composition space and the difference in the time step. Therefore, the MFIX-ISAT algorithm was modified to improve ISAT performance by calling ISAT from MFIX at a fixed time step. With the improved algorithm, ISAT reduced the computational time for chemistry drastically and the speed-up was around 46 times for silane pyrolysis.

Then two-dimensional and three-dimensional non-isothermal silane pyrolysis cases were used to test the features of ISAT. In ISAT, positive scale factors can be assigned to the independent and the dependent variables. Thus, ISAT can tabulate in terms of the normalized quantities to improve the performance. Two-dimensional non-isothermal silane pyrolysis cases with and without scaled factor were simulated. An error tolerance of 10^{-5} for the case without scaled factors was chosen to maintain the calculation accuracy. A large error tolerance $(E=10^{-2})$ can be used to maintain the agreement with DI when using scaled factors, thus reducing computational time. The maximum allowable table size was exceeded for the case without scaled factors, whereas a speedup of 4 times was obtained for the case with scaled factors. Since the values of the variables were quite different in the freeboard than in the bed, two tables were created separately to maintain accuracy and reduce computational time. One table was built for chemical reactions in the bed, which included the gas-solid phase reactions and gas phase reactions. Another table was for the freeboard and only tabulated the gas phase reactions. With the non-isothermal three-dimensional case, the speed-up was around 50 times on one processor. The total CPU time for simulations was reduced by 24% by using ISAT.

All capabilities of ISAT were tested with complex chemical reactions in a fluidized bed coal gasifier. The reactions of the coal gasifier were divided into initial stage reactions and gasification and combustion reactions. With such complex reactions, the PMSR model was tested first with the chemical reactions to provide information for optimizing parameters in ISAT. An error tolerance of $E=10^{-2}$ was necessary to maintain reasonable accuracy for fluidized bed simulations. The speed-up was around 20.5 times with PMSR reactor. A batch coal gasifier was simulated with Illinois No.6 coal using the optimized parameters obtained from PMSR simulations. DI and ISAT compared well for both dense bed and freeboard regions and the speed-up of chemistry was around 16 times with 10% reduction in total CPU time for MFIX-ISAT simulations.

It becomes possible to simulate complex multiphase flow reactors, such as biomass gasifiers, when chemistry is efficiently calculated using ISAT. Biomass has gained consideration as a promising and profitable energy source and is an excellent alternative fuel to help reduce global warming and the use of fossil fuels. However, the entrainment of carbon particles into the freeboard from the reacting bed is the main source of loss in reactor efficiency, especially for biomass reactors, since attrition and fragmentation are far more extensive for biomass. In addition, the elutriation of solid particles has to be known for designing gas cleaning equipment. As a first step to investigate a biomass gasifier, elutriation phenomenon was simulated and validated with experiments from the literature and those performed at Iowa State University.

The first experimental work for elutriation phenomenon in fluidized beds was performed by Leva (1951) and chosen as the benchmark case to test the feasibility of MFIX to accurately predict elutriation. In the simulations, the fine and coarse sand particles were solved as two individual phases. Effects of gas velocity, and fine and coarse particle diameters were investigated for elutriation rate constants above the TDH. The results of the experiments and the simulations compared well. The elutriation rate constants were proportional to approximately the 5.6th power of the gas velocity and decreased with increasing fine diameters. The diameters of coarse particles had little influence on elutriation rate constants. Experiments performed at ISU investigated the influence of gas velocity for the elutriation rate when the reactor height was below the TDH. Four different gas velocities were used and experimental results compared very well with the simulations.

Apart from the mainstream research, the effects on the use of coordinate systems and configurations to model fluidized bed reactors were tested. Since three-dimensional simulations of fluidized beds, based on the Eulerian-Eulerian model, are usually prohibitive, two-dimensional simulations are commonly used to compare with experiments. Thus, the validity of using 2D simulations to represent the true dynamics and the choice of coordinate system and configuration used to model a cylindrical reactor were tested. Three different fluidization regimes:



Figure 9.1 CFD simulations of biomass fluidized bed reactors.

bubbling, slugging and turbulent regimes, were investigated. The results indicated that a 2D Cartesian system can be used to successfully simulate and predict a bubbling regime. Caution must be exercised when using 2D simulations for other fluidized regimes.

For the future work, a laboratory-scale, bubbling fluidized bed biomass gasifier should be simulated using MFIX. Cold flows of the gasifier should be computed to test the hydrodynamics of the flow field first. The biomass gasification kinetics should then be implemented into MFIX with ISAT. The chemical kinetic models can be tested with a simple reactor using a pairwise mixing stirred reactor for validation first. For biomass, the loosely connected structures of chars after devolatilization form carbon fines by comminution such as attrition and fragmentation. Carbon comminution during fluidized bed reactions has gained growing interest because it controls the elutriation of fines, therefore, reactor efficiency and pollution emission. The particle population balance method is the starting point to account for carbon comminution phenomena. Particle size distribution due to comminution can be tracked by the direct quadrature method of moments (DQMOM) (Marchisio and Fox, 2005). DQMOM is a computationally efficient and effective method to simulate the evolving particle-size distribution in fluidized beds. Fan et al. (2004) simulated fluidized beds without reactions using MFIX with DQMOM. Therefore, DQMOM can be used in conjunction with ISAT to simulate the particle size change due to the comminution and reactions in a chemically reacting biomass reactor, as shown in Fig. 9.1.

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