Computational Fluid Dynamics Modeling and Simulations of Fluidized Bed Reactors for Chemical Looping Combustion

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Computational Fluid Dynamics Modeling and Simulations of Fluidized Bed Reactors for Chemical Looping Combustion
by
Subhodeep Banerjee

A dissertation presented to the
Graduate School of Arts & Sciences
of Washington University in
partial fulfillment of the
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of Doctor of Philosophy

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Subhodeep Banerjee

Washington University in St. Louis

August 2016
To Umma.
ABSTRACT OF THE DISSERTATION

Computational Fluid Dynamics Modeling and Simulations of Fluidized Bed Reactors for Chemical Looping Combustion

by

Subhodeep Banerjee

Doctor of Philosophy in Mechanical Engineering

Department of Mechanical Engineering and Materials Science

Washington University in St. Louis, 2016

Dr. Ramesh K. Agarwal, Chair

Chemical looping combustion (CLC) is a next generation combustion technology that shows great promise as a solution for the need of high-efficiency low-cost carbon capture from fossil fueled power plants. To realize this technology on an industrial scale, the development of high-fidelity simulations is a necessary step to develop a thorough understanding of the CLC process. Although there have been a number of experimental studies on CLC in recent years, CFD simulations have been limited in the literature.

In this dissertation, reacting flow simulations of a CLC reactor are developed using the Eulerian approach based on a laboratory-scale experiment of a dual fluidized bed CLC system. The salient features of the fluidization behavior in the air reactor and fuel reactor beds representing a riser and a bubbling bed respectively are accurately captured in the simulation. This work is one of the first 3-D simulations of a complete circulating dual fluidized bed system; it highlights the importance of conducting 3-D simulations of CLC systems and the need for more accurate empirical reaction rate data for future CLC simulations.
Simulations of the multiphase flow with chemical reactions in a spouted bed fuel reactor for coal-direct CLC are performed based on the Lagrangian particle tracking approach. The Discrete Element Method (DEM) provides the means for tracking the motion of individual metal oxide particles in the CLC system as they react with the fuel and is coupled with CFD for capturing the solid-gas multiphase hydrodynamics. The overall results of the coupled CFD-DEM simulations using Fe-based oxygen carriers reacting with gaseous CH₄ demonstrate that chemical reactions have been successfully incorporated into the CFD-DEM approach. The simulations show a strong dependence of the fluidization performance of the fuel reactor on the density of bed material and provide important insight into selecting the right oxygen carrier for the enhanced performance.

Given the high computing cost of CFD-DEM, it is necessary to develop a scaling methodology based on the principles of dynamic similarity that can be applied to expand the scope of this approach to larger CLC systems up to the industrial scale. A new scaling methodology based on the terminal velocity is proposed for spouted fluidized beds. Simulations of a laboratory-scale spouted fluidized bed are used to characterize the performance of the new scaling law in comparison with existing scaling laws in the literature. It is shown that the new model improves the accuracy of the simulation results compared to the other scaling methodologies while also providing the largest reduction in the number of particles and in turn in the computing cost.

CFD-DEM simulations are conducted of the binary particle bed associated with a coal-direct CLC system consisting of coal (represented by plastic beads) and oxygen carrier particles and validated against an experimental riser-based carbon stripper. The simulation results of the particle behavior and the separation ratio of the particles are in excellent agreement with the experiment. A credible simulation of a binary particle bed is of particular importance for understanding the details of the
fluidization behavior; the baseline simulation established in this work can be used as a tool for designing and optimizing the performance of such systems.

The simulations conducted in this dissertation provide a strong foundation for future simulations of CD-CLC systems using solid coal as fuel, considering the additional complexities associated with the changing density and diameter of the coal particles as devolatilization and gasification process occur. A complete reacting flow simulation in the CFD-DEM framework will be crucial for the successful deployment of CD-CLC technology from the laboratory scale to pilot and industrial scale projects.
Chapter 1

Introduction

The relationship between the global surface temperature of the earth and the concentration of carbon dioxide (CO₂) in the atmosphere was discovered by Arrhenius in 1896 [1]. CO₂ and other gases such as CH₄ produced from burning fossil fuels trap thermal radiation from the Sun in the Earth’s atmosphere and lead to an increase in the Earth’s surface temperature by the greenhouse effect. Following the industrial revolution at the turn of the 20th century, the world began to consume fossil fuels for energy at breakneck pace for applications ranging from energy production to transportation to water supply. The availability of these services is necessary to maintain and improve the standard of life to which the developed world has grown accustomed. However, as a result of the rapid combustion of fossil fuels, the level of CO₂ in the atmosphere has risen by almost 30% compared to the pre-industrial times and the Intergovernmental Panel on Climate Change [2] has reported that the “warming of the climate system is unequivocal” and “most of the observed increase in global average temperatures since the mid-20th century is very likely due to the observed increase in anthropogenic greenhouse gas concentrations.” This global warming is projected to cause an increase in sea levels caused by melting of the polar ice caps as well as increase in the frequency and intensity of extreme weather events. Although renewable energy is expected to account for an increasing amount of the energy supply in the future, fossil fuels will remain the dominant energy source for at least the next 25 years as shown in Figure 1.1 [3]. As a result, addressing carbon emissions from power plants has become an active area of research.
In recent years, several technologies have been demonstrated to capture CO₂ emissions from fossil fueled power plants and greatly reduce emissions into the atmosphere. These technologies can be broadly categorized as pre-combustion capture such as the integrated gas combined cycle (IGCC), post-combustion capture such as sorbent-based absorption, and oxy-fuel combustion. However, each of these technologies require a separate process to isolate CO₂ from the other gases, which consumes much of the total energy produced by the plant and can lead to a significant increase in the cost of electricity. One technology that has shown great promise for high-efficiency low-cost carbon capture is chemical looping combustion (CLC). The CLC process typically utilizes dual fluidized bed reactors—an air reactor and a fuel reactor—and a metal oxide oxygen carrier that circulates between the two reactors, as illustrated in Figure 1.2(a). Another setup for CLC that has been documented in the literature employs a single vessel with a packed bed of oxygen carrier that is alternatingly used as an air and fuel reactor via a high temperature gas switching system, shown in Figure 1.2(b). The primary advantage of CLC is that the combustion of fuel in the fuel reactor takes place in the absence of air using oxygen provided by the oxygen carrier; the flue stream from
the fuel reactor is not contaminated or diluted by other gases such as nitrogen. This provides a high-purity carbon dioxide stream available for capture at the fuel reactor outlet without the need for an energy-expensive gas separation process. The reduced oxygen carrier from the fuel reactor is pneumatically transported to the air reactor where it is re-oxidized by oxygen from air and circulated back to the fuel reactor to complete the loop.

![Diagram of chemical looping combustion system](image)

Figure 1.2. Schematic representation of a chemical looping combustion system with (a) interconnected fluidized beds, and (b) packed bed with alternating flow [4]

The only energy cost of separation associated with CLC is the cost of solid recirculation. This is considerably lower than the benchmark for pre-combustion technologies for carbon capture such as oxy-fuel combustion where the oxygen separation process can consume about 15% of the total energy. Therefore, CLC holds the answer as the next-generation combustion technology due to its potential to allow CO$_2$ capture with little to no effect on the efficiency of the power plant. Several studies on the energy and exergy of CLC systems in the literature suggest that power efficiencies greater than 50% can be achieved along with nearly complete CO$_2$ capture [5, 6, 7].
Chapter 2

Current State of the Art

The oxygen carrier used in chemical looping combustion directly affects the performance of the CLC system; a number of factors must be considered prior to its selection. The thermodynamic equilibrium for the reaction with the fuel must be such that high conversion rates are possible. The reaction rates for oxidation and reduction must be fast to avoid requiring a large amount of the oxygen carrier in the reactors. Laboratory-scale experimental studies with various oxygen carriers have identified Mn-, Fe-, Ni-, Cu-, and Co-based oxygen carriers as suitable choices for CLC operation [8, 9, 10]. However, Jerndal et al. [10] found that when Ni- and Co-based oxygen carriers are used along with solid coal fuel, the sulfur compounds in the coal may react with the metal oxide to form metal sulfides or sulfates resulting in the deactivation of the oxygen carrier particles. Ni-based carriers may also pose health hazards due to their toxicity. Of the rest, the Mn$_3$O$_4$ oxygen carrier was found to provide the highest energy output in a separate study using process simulation models developed in Aspen Plus in a previous paper [11]. Other factors in the selection of oxygen carrier include the cost as well as the physical characteristics for reactivity and fluidization. Since iron is among the cheapest and most abundant metals available on Earth, Fe-based metal oxides such as Fe$_2$O$_3$ (hematite) are well-suited for use as the oxygen carrier.

A great deal of early research in the area of chemical looping combustion focused primarily on the use of gaseous fuels such as natural gas and syngas. However, as shown in Figure 1.1, since coal is projected to remain one of the dominant fossil fuels in the near future, the use of coal for CLC has garnered significant interest in recent years. One way to utilize coal in a CLC process is to first
gasify the coal into syngas in a standalone gasifier and then inject the freshly-converted syngas into the fuel reactor. To ensure the absence of nitrogen and other gases in the syngas, the gasification must be carried out with oxygen instead of air, which requires an additional air separation unit. As such, this approach introduces the inefficiencies associated with oxy-fuel combustion and similar technologies. From the perspective of the CLC process, this scenario is identical to the one that uses gaseous fuel. The alternate approach is to inject pulverized coal directly into the fuel reactor, a process known as coal-direct chemical looping combustion (CD-CLC). The CD-CLC concept eliminates the necessity of a separate gasifier and reduces the complexity of the power plant. Within CD-CLC, two alternatives have been proposed as to how the metal oxide will participate in the coal combustion since the solid-solid reaction rate of coal with the metal oxide is negligible [12]. One option known as CD-CLC with in situ gasification, which is considered in this report, is to gasify the coal in the fuel reactor with CO₂ or H₂O as the fluidized agent and react the oxygen carrier with the products of gasification [13]. The other option, based on a patent by Lewis and Gilliland [14] and discussed in the context of chemical looping by Mattison et al. [15], is known as chemical looping with oxygen uncoupling (CLOU), which utilizes special oxygen carriers that release gaseous oxygen under the reactor conditions that can sustain the combustion of solid coal in the fuel reactor.

The work of Leion et al. [12] identified that the rate of fuel conversion in the CD-CLC process is limited by the char gasification step. The agglomeration between oxygen carrier and coal ash is another concern as it has been reported to reduce the reactivity of the metal oxide particles [16]. These concerns are addressed by utilizing a spouted fluidized bed for both reactors with relatively large diameter particles, unlike in CLC using gaseous fuels that can use a bubbling or fast fluidized bed for the fuel reactor. The larger particles correspond to Group D or spoutable particles according
to Geldart’s powder classification [17]. In a spouted fluidized bed, a high velocity jet of pulverized coal and the fluidizing agent is injected at the center of the fluidized bed to induce strong circulation rates for the solid particles and enhance the solid-gas mixing. The increased friction from the mixing of solids can also serve to slough off the ash build-up on the metal oxide particles and restore reactivity [18].

All of the previous work discussed so far in this report involved laboratory-scale experiments of CLC systems. Setting up and executing a laboratory experiment can be an expensive and laborious process. On the other hand, computational fluid dynamics (CFD) provides an efficient means to analyze the performance of a CLC system and characterize the fluid mechanics and chemical kinetics in the system. However, although laboratory-scale studies of CLC with various experimental setups are widespread in the literature, numerical studies using CFD have been limited. Initial CFD studies in the field demonstrated the capability of computational methods to model a multiphase gas-solid system and were not based on any particular experiment [19, 20]. Later, the work of Mahalatkar et al. [21, 22] based on a single reactor setup similar to Figure 1.2(b) showed that CFD simulation is able to match the reaction mechanics inside a CLC fuel reactor with reasonable accuracy. However, the single reactor setup “cannot be operated with solid fuels and the design and operation of the hot gas switching system is problematic” [4]. To design a CLC system for operation using solid coal given its likelihood to remain a dominant fossil fuel in the near future, the use of the dual fluidized bed setup shown in Figure 1.1(a) is necessary. The single reactor simulations that exist in literature do not provide any information about the circulation of oxygen carrier inside a dual fluidized bed setup. Therefore, it is important to establish a credible CFD simulation based on the interconnected fluidized bed setup for CLC. To accurately model the fluidized bed configuration with particulate metal oxides and fuel present, it is critical to accurately
capture the solid circulation and separation as a result of the solid-gas two-way coupling and the solid-solid interaction. Numerical modeling of multiphase flows involving a granular solid and a gas of the kind seen inside a CLC fuel reactor can be achieved with different levels of accuracy with very differing computational costs depending on the modeling approach, which can be broadly categorized as either Eulerian or Lagrangian.

The Eulerian or multi-fluid model circumvents the high computational demand of the particle-based models while retaining fidelity. In the multi-fluid models, the solid phase is also considered as a continuum fluid and particle variables such as mass, velocity, temperature, etc. are averaged over a region that is large compared to the particle size. As such, this approach only accounts for the bulk behavior of the solid phase. Constitutive equations for the solid phase pressure and solid phase viscosity, which are required to model the interactions between the solid and the gas phases, are provided by the kinetic theory of granular flow. The kinetic theory of granular flow is an extension of the classical kinetic gas theory that includes the inelastic particle-particle interactions [23, 24]. The multi-fluid models are a simpler approach to investigate the fluid properties compared to the particle-based method, and have been successfully implemented in a wide range of fluidization applications such as bubbling fluidized beds [25], particle mixing [26], down-flow reactors [27] and spouted beds [28]. Jung and Gamwo [19] were the first to apply the multi-fluid approach for modeling the CLC process. However, the drawback of the Eulerian multi-fluid model is that the exact particle dynamics in the system cannot be determined and optimized.

In the Lagrangian particle-based model or discrete element method (DEM), the trajectory of each individual particle is resolved based on a force balance calculation for the particle. The particle tracking is coupled with CFD solution for the fluid phase by considering the interaction between the particles and the fluid separately for each particle. DEM can model the properties such as
temperature, composition, position, and velocity with high accuracy, limited only by the specifics of the particle collision parameters employed in the model. Since the number of particle collisions increase drastically with an increase in the number of particles, the number of particles in a DEM simulation presented here is constrained to around $10^5$ because of limitation of available computational capability; this number can be increased if greater computing power is available. The high computational cost is the reason behind the scarcity of particle-based models for CLC simulation in the literature to date. However, cold-flow simulations using the coupled CFD/DEM model have proven capable in accurately matching the particle dynamics of various laboratory scale fluidized bed experiments using relatively large Geldart Group D particles [29, 30]. However, no work has been done in the literature on integrating the chemical reactions into a CFD/DEM model. All the modeling work that has been done on the chemical reactions aspect of CD-CLC has focused either on Eulerian simulations [21, 22] or on process simulations from an energy balance perspective using software such as Aspen Plus [31, 32, 33]. It is crucial to combine the solid particle dynamics and chemical reactions into one credible model for the whole CD-CLC fuel reactor that can be used to investigate various aspects of CD-CLC including reactor design, inlet jet velocity, and the physical properties of the oxygen carrier in order to achieve an optimized design in the future.
Chapter 3

Eulerian Approach to Numerical Simulation

All modeling work in this dissertation is performed using the commercial CFD simulation package ANSYS Fluent, release version 14.5 [34, 35]. Since the flow inside a CLC fuel reactor is chemically active with heat transfer, all the equations of fluid dynamics—the continuity equation, the Navier-Stokes momentum equations, and the energy equation—are required in order to capture the flow field. The solid phase is approximated as an Eulerian fluid phase whereby particle variables such as mass, velocity, temperature, etc. are averaged over a region that is large compared to the particle size. Thus, the Eulerian approach only accounts for the bulk behavior of the solids. Constitutive equations for the solid phase pressure and viscosity are required to model the interactions between the solid and gas phases. These are provided by the kinetic theory of granular flow, which is an extension of the classical kinetic gas theory that includes inelastic particle/particle interactions [23, 24]. The Eulerian framework for modeling a multiphase flow involving a granular solid and a gas has been in use for a few decades and has proven reliable in capturing the experimental behavior. Details of the equations used to compute the fluid and solid motion are provided in the following subsection.

3.1 Modeling Approach for Eulerian Simulation

For multiphase simulations using the Eulerian approach, the standard equations of fluid motion are slightly modified to account for the presence of additional phases by including the porosity $\alpha$ defined as the volume fraction of the respective phase in the computational cell where the equations are applied [35]. The continuity equation for phase $q$ is given as
\[ \frac{\partial}{\partial t} (\alpha_q \rho_q) + \nabla \cdot (\alpha_q \rho_q \mathbf{u}_q) = \sum (\dot{m}_{pq} - \dot{m}_{qp}) \] (1)

where \( \dot{m}_{pq} \) is the mass transfer rate from the \( p \)th phase to the \( q \)th phase. Each phase (gas or solid) consists of a number of species. A transport equation is solved for each species,

\[ \frac{\partial}{\partial t} (\alpha_q \rho_q Y_{iq}) + \nabla \cdot (\alpha_q \rho_q \mathbf{u}_q Y_{iq}) = \sum (\dot{m}_{ij}^{qp} - \dot{m}_{ji}^{pq}) \] (2)

where \( Y_{iq} \) is the mass fraction of the species \( i \) in the \( q \)th phase and \( \dot{m}_{ij}^{qp} \) is the mass transfer rate from the \( j \)th species of the \( p \)th phase to the \( i \)th species in the \( q \)th phase. In this work, one gas phase and one solid phase is considered, corresponding to the fuel-gas mixture and the oxygen carrier respectively.

The momentum equation for the gas phase is given as

\[ \frac{\partial}{\partial t} (\alpha_g \rho_g \mathbf{u}_g) + \nabla \cdot (\alpha_g \rho_g \mathbf{u}_g \mathbf{u}_g) = -\alpha_g \nabla p + \nabla \cdot \mathbf{\tau} + \alpha_g \rho_g \mathbf{g} + \sum (R_{sg} + \dot{m}_{sg} \mathbf{u}_{sg} - \dot{m}_{gs} \mathbf{u}_{gs}) \] (3)

where the terms in the summation are source terms added to the standard form of the Navier-Stokes momentum equations to account for the momentum transfer between the solid phase and the gas phase. Specifically, \( R_{sg} = \beta_{sg} (\mathbf{u}_s - \mathbf{u}_g) \) is the momentum transfer due to interphase drag and the other terms are due to the transfer of mass. The momentum equation for the solid phase follows from the momentum equation for the gas phase with the source term for interphase drag being equal but opposite.

\[ \frac{\partial}{\partial t} (\alpha_s \rho_s \mathbf{u}_s) + \nabla \cdot (\alpha_s \rho_s \mathbf{u}_s \mathbf{u}_s) = -\alpha_s \nabla p + \nabla \cdot \mathbf{\tau}_s + \alpha_s \rho_s \mathbf{g} + \sum (R_{rs} + \dot{m}_{rs} \mathbf{u}_{rs} - \dot{m}_{sr} \mathbf{u}_{sr}) \] (4)

For the flow conditions in a fuel reactor, the gas can be considered as an incompressible fluid. The fluid stress tensor is simply the Cauchy stress tensor with zero bulk viscosity.
\[ \bar{\tau}_g = \alpha_g \mu_g (\nabla \mathbf{u}_g + \nabla \mathbf{u}_g^T) \]  \hspace{1cm} (5)

On the other hand, the granular solid stress tensor considers all terms in the Cauchy stress tensor,

\[ \bar{\tau}_s = -p_s \bar{I} + \alpha_s \mu_s (\nabla \mathbf{u}_s + \nabla \mathbf{u}_s^T) + \alpha_s \lambda_s (\nabla \cdot \mathbf{u}_s) \bar{I} \]  \hspace{1cm} (6)

where \( p_s \) is the solids pressure, \( \mu_s \) is the granular viscosity, and \( \lambda_s \) is the granular bulk viscosity. The definition of these terms and the interphase exchange coefficient \( \beta_{sg} \) provide the basis for the Eulerian approach for multiphase flow simulation. The solids pressure and granular bulk viscosity are defined according to Lun et al. [36]; the granular viscosity is according to Gidaspow [37]. The Gidaspow model is well-suited for fluidized bed simulations that include a range of solid loadings from dilute to densely packed because it accounts for the differences in the solid-gas interaction behavior in the dilute and dense regions by switching between the drag prediction of the Ergun equation [38] and the drag model of Wen and Yu [39] based on the solids fraction \( \alpha_s \). For \( \alpha_s > 0.8 \), the Gidaspow model for the exchange coefficient \( \beta_{sg} \) gives

\[ \beta_{sg} = \frac{3}{4} \frac{C_D}{d_s} \frac{\alpha_s \alpha_g \rho_g |\mathbf{u}_s - \mathbf{u}_g|}{\alpha_g^{-2.65}} \]  \hspace{1cm} (7)

Conversely, for \( \alpha_s \leq 0.8 \),

\[ \beta_{sg} = 150 \frac{\alpha_s (1 - \alpha_g) \mu_g}{\alpha_g d_s^2} + 1.75 \frac{\rho_g \alpha_s |\mathbf{u}_s - \mathbf{u}_g|}{d_s} \]  \hspace{1cm} (8)

where \( d_s \) is the diameter of the solid particles and \( \text{Re}_s \) is the Reynolds number based on \( d_s \).

Finally, the energy equation for phase \( q \) is expressed in terms of the enthalpy as

\[ \frac{\partial}{\partial t} (\alpha_q \rho_q h_q) + \nabla \cdot (\alpha_q \mathbf{u}_q h_q) = \alpha_q \frac{\partial p}{\partial t} + \nabla \cdot (\bar{\tau}_q \cdot \mathbf{u}_q) - \nabla \cdot \mathbf{q}_q + S_q + \sum Q_{pq} \]  \hspace{1cm} (9)
where \( h_q \) and \( q_q \) are the specific enthalpy and heat flux of phase \( q \) respectively. As with the continuity and momentum equations, source terms are implemented to account for the transfer of enthalpy between phases. In particular, \( S_q \) is the enthalpy source due to chemical reaction and \( Q_{pq} \) is the heat transfer from the \( p \)th phase to the \( q \)th phase. The interphase heat transfer is modeled based on Gunn [40].

### 3.2 Eulerian Simulation of the Experimental CLC Reactor of Abad et al. [41]

In this section, the laboratory scale experiment of Abad et al. [41] is used as a basis to perform a detailed CFD simulation of a CLC system using the Eulerian multi-fluid approach. It is one of the first CFD models of a complete circulating dual fluidized bed setup. The fluidization behavior in both air and fuel reactor beds and the circulation of the oxygen carrier between the beds is investigated and compared with the experiment. Chemical reactions in the fuel reactor are also considered and the CFD data is validated against the outlet concentrations of various flue gases.

#### 3.2.1 Description of Experimental Setup

The experiment uses the two-compartment fluidized bed design proposed by Chong et al. [42] and further investigated by Yang et al. [43]; the experimental setup is shown in Figure 3.1. Dimensions and additional details can be found in Abad et al. [41]. The oxygen carrier particles in the air reactor are oxidized in the presence of air; the fluidizing velocity is greater than the terminal velocity of the particles and carries the particles upwards. The flow then undergoes a sudden expansion in the particle separator at the top of the reactor, which causes the particles to fall back down into the down-comer and enter the fuel reactor.
Figure 3.1. Sketch of experimental reactor: (1) air reactor, (2) down-comer, (3) fuel reactor, (4) slot, (5) gas distributor plate, (6) wind box, (7) reactor part, (8) particle separator, (9) leaning wall. The symbols (x) and (o) indicate fluidization in the down-comer and slot.

The experiment used a Fe-based oxygen carrier consisting of 60% Fe$_2$O$_3$ by mass and 40% Al$_2$O$_3$. The analysis of several metal oxides and alloys showed that this oxygen carrier provides excellent reactivity for use in CLC and its hardness and resistance to agglomeration is ideal for fluidized bed operation [44]. The Al$_2$O$_3$ is inert and acts as a porous support providing a higher surface area for reaction. The particular batch of oxygen carrier used by Abad et al. [41] was sintered at 1100ºC and is designated as F6A1100. The gaseous fuels used in the experiment are natural gas, consisting primarily of CH$_4$, and syngas, simulated by a mixture of 50% CO and 50% H$_2$. The fluidizing velocity in the fuel reactor is below the terminal velocity of the particles, hence a bubbling bed behavior is exhibited. Therefore, the particles do not reach the particle separator in the fuel reactor. The pressure in the fuel reactor is controlled via a water trap connected to the flue stream of the reactor to ensure minimal gas leakage between the fuel reactor and the air reactor through the down-comer and slot. The flue streams from both reactors are led to a gas analyzer where the concentrations of various gases are measured.
3.2.2 Chemical Reaction Scheme and Rates

The metal oxide reduction reactions used in the simulation are

\[
3 \text{Fe}_2\text{O}_3 + \text{CO} \rightarrow 2 \text{Fe}_3\text{O}_4 + \text{CO}_2 \quad (10)
\]

\[
3 \text{Fe}_2\text{O}_3 + \text{H}_2 \rightarrow 2 \text{Fe}_3\text{O}_4 + \text{H}_2\text{O} \quad (11)
\]

Exact reaction rates for the reduction of F6A1100 with CO and H\(_2\) are not available in the literature; the reaction rates are assumed to be the same as the reduction rates for hematite (pure Fe\(_2\)O\(_3\)) with CO and H\(_2\) obtained from the experimental work of Mattisson et al. [45] and further developed by Mahalatkar et al. [22] for the simulation of chemical reactions in a single reactor setup with solid fuel. Based on these papers, the reaction rates \(\dot{m}\) (in kg/s per cell volume or kg/(m\(^3\)-s)) of the fuel gases with iron oxide are given by

\[
\dot{m}_{\text{H}_2} = \frac{k_{\text{H}_2}R_o}{2MW_{\text{O}_2}} \rho_{\text{avg}} \alpha_s \left(Y_{\text{Fe}_2\text{O}_3} + Y_{\text{Fe}_3\text{O}_4} \frac{\nu_{\text{Fe}_2\text{O}_3}MW_{\text{Fe}_2\text{O}_3}}{\nu_{\text{Fe}_3\text{O}_4}MW_{\text{Fe}_3\text{O}_4}} \right) (1 - X)^{2/3}MW_{\text{H}_2} \quad (12)
\]

and

\[
\dot{m}_{\text{CO}} = \frac{k_{\text{CO}}R_o}{2MW_{\text{O}_2}} \rho_{\text{avg}} \alpha_s \left(Y_{\text{Fe}_2\text{O}_3} + Y_{\text{Fe}_3\text{O}_4} \frac{\nu_{\text{Fe}_2\text{O}_3}MW_{\text{Fe}_2\text{O}_3}}{\nu_{\text{Fe}_3\text{O}_4}MW_{\text{Fe}_3\text{O}_4}} \right) (1 - X)^{2/3}MW_{\text{CO}} \quad (13)
\]

where \(k\) is the nominal reaction rate based on the Arrhenius rate, \(R_o\) is the oxygen carrying capacity, \(MW\) is the molecular weight (in kg/kmol), \(Y\) is the mass fraction, \(\nu\) is the stoichiometric coefficient, and \(X\) is the conversion fraction based on the fully reduced state; in each case, the subscript identifies the species under consideration. More details of the reaction rate derivation can be found in Mahalatkar et al. [22]. The reaction rates identified in Eqs. (12) and (13) are implemented into the numerical simulation through separate user-defined functions.
3.2.3 Two-Dimensional Simulation of Abad et al. Experiment

The simple rectilinear geometry of the reactor used in the experiment of Abad et al. [41] can be easily modeled using a 2-D setup. The absence of cylindrical elements such as a cyclonic separator suggests that the introduction of swirl velocities and other 3-D effects will be minimal. The numerical grid used in the CFD simulation is based on the experimental setup shown in Figure 3.1 with few changes. Since the simulation assumes a 2-D domain, the expansion in the particle separator is modified to take place in the plane of the reactor. Since the expansion takes place downstream of the circulating bed, it is expected that the bed behavior remains unaffected. The leaning wall and the wind-box are also removed and the fluidizing gas is introduced directly at the bottom of the air and fuel reactors. The resulting geometry is meshed with a relatively fine grid in the lower regions of the geometry where the solid circulation occurs (particularly in the slot region) and a coarser grid in the upper regions where the expansion takes place. This ensures that an accurate solution for the fluidization behavior is obtained while also limiting the computational cost. The final CFD mesh is shown in Figure 3.2.

The experimental reactor of Abad et al. [41] was operated for 60 hours with both natural gas and syngas without replacing the oxygen carrier particles or adding new material. Given the time taken to complete 20 seconds of simulation on a Dell workstation with a quad-core Intel Xeon central processing unit (approximately 48 hours), the complete reactor simulation of 60 hours is beyond the scope of CFD at this time. Instead, the initial batch processing results of Abad et al. [41] are used to validate the CFD simulation in the present work. For these batch operations, which last less than one minute, the initial oxygen carrier mass in the fuel reactor is sufficient for reacting with all the incoming fuel, so the fuel conversion is not affected by the oxygen carrier re-oxidation in the air reactor. The CFD simulation is thus considerably simplified by setting the fluidization gas in the
air reactor to an inert gas (nitrogen in this case). Of the two gaseous fuels used in the experiment, only syngas has been considered in the current work because the chemical kinetics for the reaction of Fe$_2$O$_3$ with the non-methane components of natural gas are not available.

The oxygen carrier used in the experiment has an apparent density of 2,150 kg/m$^3$ and porosity of 0.56 with a diameter of 90–212 μm; the average value of 150 μm is used in the simulation. The initial bed height is 10 cm, corresponding to a bed mass of 180 g. The reactors are set to atmospheric pressure and the gage pressures at the outlets are set at zero. The pressure differential between the reactors, controlled by a water trap in the experiment to minimize leakage, was not implemented in the simulation because the data was not available. It is expected that the pressure differential is a secondary mechanism and the dense solid packing in the down-comer and slot will be sufficient to keep the leakage to a minimal. Lastly, the temperature for the simulation was set at 1123 K, in line with the reference condition specified from the experiment. The numerical parameters used in the CFD simulation are summarized in Table 3.1. It should be noted that the secondary phase mass fraction has been set to zero at both fuel and air reactor inlets, i.e., no new oxygen carrier is added.
Table 3.1. Key modeling parameters for CFD simulation of Abad et al. experiment [19]

<table>
<thead>
<tr>
<th>Primary phase</th>
<th>Fuel-gas mixture</th>
</tr>
</thead>
<tbody>
<tr>
<td>Secondary phase</td>
<td>Oxygen carrier (F6AL1100)</td>
</tr>
<tr>
<td>Average particle diameter</td>
<td>150 μm</td>
</tr>
<tr>
<td>Average particle density</td>
<td>2150 kg/m³</td>
</tr>
<tr>
<td>Initial bed mass</td>
<td>~180 g</td>
</tr>
<tr>
<td>Fluidizing gas composition in fuel reactor</td>
<td>50% CO, 50% H₂</td>
</tr>
<tr>
<td>Fluidizing gas composition in air reactor</td>
<td>100% N₂</td>
</tr>
<tr>
<td>Inlet boundary condition in fuel reactor</td>
<td>Velocity inlet with velocity 0.1 m/s</td>
</tr>
<tr>
<td>Inlet boundary condition in air reactor</td>
<td>Velocity inlet with velocity 0.5 m/s</td>
</tr>
<tr>
<td>Outlet boundary condition in fuel reactor</td>
<td>Pressure outlet at atmospheric pressure</td>
</tr>
<tr>
<td>Outlet boundary condition in air reactor</td>
<td>Pressure outlet at atmospheric pressure</td>
</tr>
<tr>
<td>Operating temperature</td>
<td>1123 K</td>
</tr>
<tr>
<td>Solids pressure</td>
<td>Lun et al. [36]</td>
</tr>
<tr>
<td>Granular bulk viscosity</td>
<td>Lun et al. [36]</td>
</tr>
<tr>
<td>Granular viscosity</td>
<td>Gidaspow [37]</td>
</tr>
<tr>
<td>Drag law</td>
<td>Gidaspow [37]</td>
</tr>
<tr>
<td>Heat transfer coefficient</td>
<td>Gunn [40]</td>
</tr>
<tr>
<td>Numerical scheme</td>
<td>Phase-coupled SIMPLE</td>
</tr>
<tr>
<td>Time step size</td>
<td>0.0005 s</td>
</tr>
<tr>
<td>Iterations per time step</td>
<td>20</td>
</tr>
</tbody>
</table>

The simulation was run for approximately 20 seconds. Figure 3.3 shows the instantaneous contour plots of the solid volume fraction during the initial development of solids flow in the dual fluidized bed reactor system for the first one second. Initially, the oxygen carrier particles in the air reactor are...
lifted by the fluidizing velocity until they reach the particle separator. The reduction in flow velocity due to the expansion in the particle separator leads the particles to drop into the air reactor bed and the down-comer; this leads to a densely packed bed in the down-comer nearing the packing limit. The dense packing is beneficial because it seals the passage and minimizes leakage of nitrogen from the air reactor to the fuel reactor, ensuring that the flue stream of the fuel reactor is a high purity CO$_2$ stream uncontaminated by nitrogen ready for capture. It can also be seen that the particles in the fuel reactor are not significantly lifted by the flow because of the lower velocity of the fluidized gas; the bed configuration in the fuel reactor represents a bubbling fluidized bed. Although it is not evident from Figure 3.3, inspection of the solid phase velocity vectors indicates that the motion of solid particles in the slot or loop-seal is in the correct direction, i.e., from the fuel reactor to the air reactor. In this way, the salient features of fluidization behavior in the dual fluidized bed reactor used by Abad et al. [41] are completely captured by the CFD simulation of the reactor. However, the exact distribution of particles in the experiment could not be observed given the solid walls of the reactor, therefore a direct comparison cannot be made.

The contour plots of the solid volume fraction at subsequent times (not shown) indicate that the flow of solids in the reactor system stabilizes after around one second and continues in the same fashion for the remainder of the simulation. On the other hand, the contours of mass fraction of CO$_2$ given in Figure 3.4 for the first five seconds of simulation show that the development of the flow in the gas phase takes longer. The simulation is initialized with nitrogen in both reactor beds; this represents the nitrogen cycling performed in the experiment after each reaction period. As the syngas is injected into the fuel reactor, it starts to react with the Fe$_2$O$_3$ in the oxygen carrier and reduces it to Fe$_3$O$_4$. The CO$_2$ produced by the reaction forms a plume that first reaches the fuel reactor outlet just after two seconds. As the simulation continues, the plume becomes stronger as
more and more CO₂ is produced by the reduction of Fe₂O₃. It is noted that the mass fraction of H₂O follows the exact same contours as the mass fraction of CO₂ shown in Figure 3.4, albeit with different values. Figure 3.4 confirms that the dense packing in the down-comer and slot regions limits the leakage of CO₂ into the air reactor to a very small amount.

![Figure 3.3. Contours of solid volume fraction for the first second of 2-D simulation showing the initial development of solids flow inside the dual fluidized bed system; the maximum value of 0.63 represents the solids packing limit](image1)

![Figure 3.4. Contours of CO₂ mass fraction for the first five seconds of 2-D simulation showing the vortex pattern seen during the initial development of gas flow inside the fuel reactor](image2)
An interesting observation from Figure 3.4 is that the CO₂ plume does not diffuse into the nitrogen present in the reactor from the initialization step; instead, it travels in a vortex pattern towards the fuel reactor outlet. This is because diffusion of gases is an inherently 3-D process and cannot be captured accurately in a 2-D simulation. Consequently, pockets of reversed flow begin to develop in the fuel reactor. This is reflected in Figure 3.5, which shows large fluctuations in the concentrations of CO₂ and H₂O measured at the fuel reactor outlet as the pockets of reversed flow lead to alternating CO₂/H₂O-rich and CO₂/H₂O-lean gas at the outlet surface. As the simulation continues, the vortex pattern dissipates and the pockets of reversed flow coalesce into a single, stable plume of reversed flow, which can be clearly seen in Figure 3.6. Although this stabilizes the concentrations of CO₂ and H₂O at the fuel reactor outlet, their values are much lower than the expected values from Abad et al.’s experiments (indicated on Figure 3.5). The discrepancy can be explained by the plume of reversed flow consisting of a large amount of nitrogen at the fuel reactor outlet greatly lowering the mass fractions measurements for the other gases.

Figure 3.5. Mass fractions of CO₂ and H₂O at the fuel reactor outlet for the 2-D simulation showing fluctuations caused by the vortex pattern seen in Figure 3.4 during initial development of flow.
3.2.4 Three-Dimensional Simulation of Abad et al. Experiment

Although the 2-D simulation presented in the previous section was able to capture the salient features of the fluidization behavior in the dual fluidized bed system, it was unable to produce the expected concentrations of CO₂ and H₂O in the fuel reactor because of the inadequacy of the 2-D simulation in modeling the gaseous diffusion. In this section, a 3-D simulation of the experiment of Abad et al. is conducted to obtain a closer fit of the reaction results between the simulation and experiment. Unlike the 2-D domain, which used a modified version of the reactor geometry with the particle separator in plane, the 3-D computational domain is an exact representation of the geometry of Abad et al. [41]. Similar to the 2-D case, a structured mesh is used with a relatively fine grid in the lower part of the reactor and a coarser grid in the upper regions. The mesh used for the 3-D simulations is shown in Figure 3.7. The initial solids loading in the bed is about 300 g, of which 110 g is in the fuel reactor, in line with the experiment. All numerical parameters for the simulation are kept the same as in Table 3.1. The rates for the reactions given in Eqs. (10) and (11) are identical to those used for the 2-D case.
The 3-D simulation was run for 30 seconds. Since the solids loading in the current simulation has been increased from the 2-D case to match the experimental value, it takes slightly longer to achieve the pressure buildup required for fluidization; the solid particles begin to display fluidization behavior starting at around 5 seconds. Once the fluidization starts, despite the difference in the configuration of the particle separator (in plane versus out of plane), there are no significant differences in the fluidization behavior of the solids between the 2-D and 3-D simulations. This is expected because there were no sources of swirl, etc. in the experiment that would lead to a different fluidization behavior in 3-D. Indeed, this result validates the reasoning behind conducting a 2-D simulation in the first place. Thus, both 2-D and 3-D simulations can accurately capture all the different fluidization regimes in the experimental dual fluidized bed reactor of Abad et al. [41].

On the other hand, there are stark differences between the 2-D and the 3-D simulations when the development of gas flow is investigated. The contours of the mass fraction of CO$_2$ for the 3-D simulation are shown in Figure 3.8. As expected, there is greater diffusion in the 3-D case. The local mass fraction of CO$_2$ at the base of the bed, where the injected CO first comes into contact
with the Fe\(_2\)O\(_3\) and begins to react is around 15\%, the same as in the 2-D simulation. However, owing to the increased diffusion in 3-D, the CO\(_2\) spreads through the fuel reactor more homogeneously as it travels towards the fuel reactor outlet; the vortex patterns seen in the 2-D case are absent. The contours of the mass fraction of H\(_2\)O display the same characteristics. The quantitative effects of this can be observed from the plot of the mass fractions of CO\(_2\) and H\(_2\)O at the fuel reactor outlet given in Figure 3.9. The mass fractions of both CO\(_2\) and H\(_2\)O are initially lower than in the 2-D case. This is because these gases now have to diffuse through the nitrogen present in the fuel reactor instead of displacing it and thus reach the outlet more slowly. The large fluctuations in the outlet mass fraction caused by pockets of reversed flow in the 2-D case are eliminated. Since reversed flow does not develop in the current simulation, the mass fractions keep increasing as the simulation progresses and more and more CO\(_2\) and H\(_2\)O are produced. By 20 seconds, the mass fractions of both CO\(_2\) and H\(_2\)O have exceeded their stagnation values from the 2-D simulation (shown by dotted lines in Figure 3.9). By 30 seconds, the mass fraction of H\(_2\)O reaches the expected value from the batch experiments of Abad et al. [41]. However, it is noted that although the final outlet mass fraction of CO\(_2\) after 30 seconds is higher than the 2-D case, it does not reach the experimental value.
Figure 3.8. Contours of CO$_2$ mass fraction for the first ten seconds of 3-D simulation showing the increased diffusion and absence of the vortex pattern compared to the 2-D case

Figure 3.9. Mass fractions of CO$_2$ and H$_2$O at the fuel reactor outlet for 3-D simulation

The 3-D simulation shows a significant improvement in the mass fraction measurements of the flue gases at the fuel reactor outlet. However, there is still some discrepancy in the mass fraction of CO$_2$, which may be due to various external factors. In Abad et al.’s experiment [41], the gas streams from the air and fuel reactor outlets were pipelined via an electric cooler into the gas analyzer. It is well
established that significant apparent diffusion can occur in gases when they travel through pipes [46]. Thus, it is reasonable to expect that the concentrations measured at the gas analyzer may be different from the concentrations present right at the fuel reactor outlet, which is what is recorded in the simulation. It should also be noted that the reaction rate kinetics used in the simulation were based on the experimental study of Mattisson et al. [45] using pure Fe$_2$O$_3$ whereas the oxygen carrier used in the experiment was F6A1100. One of the reasons F6A1100 is preferred over Fe$_2$O$_3$ as the oxygen carrier for CLC operation is its improved reactivity [44], caused by an increase in apparent surface area due to the presence of the porous Al$_2$O$_3$. Given the improved reactivity, it stands to reason that the experiment shows a higher concentration of the reaction products compared to the simulation. Further research is required to determine more accurate empirical formulas for the reduction of F6A1100 specifically to improve the accuracy of the results of the CFD simulation.

### 3.2.5 Summary and Conclusion

CFD simulation for a complete circulating dual fluidized bed system has been developed for chemical looping combustion based on the 300 W laboratory-scale experiment of Abad et al. [41]. The oxygen carrier is modeled as an Eulerian fluid phase based on the kinetic theory of granular flow. Chemical reactions in the fuel reactor for the reduction of the oxygen carrier are implemented in the simulation based on available reaction rate data for the reduction of hematite. The CFD model has been able to accurately capture the salient features of the different fluidization regimes in the air reactor, the fuel reactor beds, and the down-comer. This work is one of the first CFD models of a complete circulating reactor system and demonstrates that CFD can provide an effective alternative approach to understanding the fluidization behavior in such a system.
The results of this study highlight the importance of capturing the diffusion of gases in a CLC reactor in ensuring that accurate results are obtained for the chemical reactions. Since diffusion is an inherently 3-D process, the shortcomings of the 2-D simulation for a CLC reactor were exposed. On the other hand, the 3-D model displayed significantly improved results for the outlet concentrations of the gases produced by the reduction of metal oxide in the fuel reactor compared to the experiment, although some discrepancy was still present in the case of CO$_2$. It is expected that the simulation accuracy can be improved significantly if more accurate empirical reaction rate data becomes available for the specific oxygen carrier and fuel considered.
Chapter 4

Lagrangian Approach to Numerical Simulation

The Eulerian two-fluid model can accurately capture the solid phase fluidization behavior in a dual fluidized bed reactor for CLC, as shown in Chapter 3. The experiment of Abad et al. [41] used as the bases for the simulation in Chapter 3 used a bubbling fluidized bed in the fuel reactor to reduce the Fe-based oxygen carrier in the presence of gaseous fuel. However, as previously mentioned, the current focus in the area of CLC is to develop a CLC system that works well with solid coal fuel. For real world application of coal-direct CLC, the implementation of a spouted fluidized bed fuel reactor offers several technical benefits. First, the introduction of high velocity jet is likely to create strong mixing of solids and gas avoiding loss of reactivity due to the ash agglomeration with the oxygen carrier [16]. Second, the oxygen carrier needs to be formed into particles with a relatively large diameter corresponding to Geldart Group D particles [17] to achieve a spoutable bed; the relatively large diameter of oxygen carrier particles compared to the coal and ash particles is likely to result in easier ash separation from the recirculating oxygen carrier. A significant differentiation of a spouted fluidized bed from others (e.g. a bubbling fluidized bed) is the intense particle-particle and particle-wall collisions. The work of Gryczka et al. [28] with 2 mm diameter particles has suggested that an accurate numerical representation of particle dynamics is not likely to be achieved for spouted beds using the granular solid phase approximation of the Eulerian approach due to “the inadequacies of the continuum model.” The inaccuracy arises from the non-
physical closure terms used in the Eulerian model such as the frictional solids viscosity or the solids pressure based on the kinetic theory of granular flow. To address these shortcomings, the Lagrangian particle-based approach coupled with CFD simulation of the fluid phase known as the coupled CFD/DEM model has been introduced in recent literature.

4.1 Modeling Approach for Lagrangian Simulation

The equations for mass and momentum conservation for the fluid phase are identical to those used in the Eulerian model given in Eqs. (2) and (3) with the exception that the source term in Eq. (3) for the solid-gas momentum exchange term, $R_{sg}$, is obtained from the average of the drag forces acting on all the discrete particles in a given computational cell. The shear stress term in the momentum equation is given in Eq. (5). Since the flow inside the CD-CLC fuel reactor is chemically active with heat transfer, the energy equation must be applied. The equation for the conservation of energy for the fluid phase can be expressed in terms of the internal energy as

$$\frac{\partial}{\partial t}(\alpha_f \rho_f E_f) + \nabla \cdot (\alpha_f u_f (\rho_f E_f + p_f)) = \nabla \cdot \left( k_f \nabla T_f - \sum h_j J_j + (\tau_f \cdot u_f) \right) + S_h \quad (14)$$

where $\rho_f, u_f, p_f, E$, and $T$ are the density, velocity, pressure, internal energy, and temperature of the fluid respectively, $g$ is the acceleration due to gravity, $k$ is the thermal conductivity, and $h_j$ and $J_j$ are the enthalpy and diffusion flux of species $j$. The source term, $S_h$, in Eq. (14) captures the net heat flux due to heat transfer from the solid to the gas phase due to the chemical reactions. Similar to the source term for momentum, it is calculated based on the average of all the discrete particles in the computational cell where the equation is applied.

The trajectory of each particle is computed by integrating the force balance on the particle, which can be written in the Lagrangian frame per unit particle mass as
\[
\frac{\partial u_p}{\partial t} = g \left( \frac{\rho_f - \rho_p}{\rho_p} \right) + F_D (u_f - u_p) + F_{col}
\]  

(15)

where the subscript \( p \) denotes an individual particle. The terms on the right hand side of Eq. (15) account for the gravitational and buoyant forces, the drag force, and an additional force due to particle-particle or particle-wall collisions. Forces such as the virtual mass force and pressure gradient force can be neglected for gas-solid flows given \( \rho_p \) far exceeds \( \rho_f \). The net drag coefficient \( F_D \) is given by

\[
F_D = \frac{18 \mu_f C_D Re_p}{\rho_p d_p^2 24}
\]  

(16)

where \( d_p \) is particle diameter, \( C_D \) is the particle drag coefficient, and \( Re_p \) is the Reynolds number based on the particle diameter defined as

\[
Re_p = \frac{\rho_f d_p |u_f - u_p|}{\mu_f}
\]  

(17)

The drag coefficient can be modeled using various empirical relations. The drag law proposed by Syamlal and O’Brien [47] is selected for the spouted fluidized beds simulations. The Syamlal-O’Brien drag law is a good choice because it uses a correction based on the terminal velocity of the particle, which is the minimum velocity that is large enough to lift the particle out of the bed and is an important parameter for characterizing a spouted bed. The Syamlal O’Brien drag law defines

\[
C_D = \left( 0.63 + \frac{4.8}{\sqrt{Re_p/v_{r,p}}} \right)^2
\]  

(18)

In Eq. (18), \( v_{r,p} \) is the terminal velocity correction for the particulate phase given by
\[ v_{r,p} = 0.5 \left( A - 0.06 \text{Re}_p + \sqrt{(0.06 \text{Re}_p)^2 + 0.12 \text{Re}_p(2B - A) + A^2} \right) \]  

(19)

where

\[ A = \alpha_f^{4.14} \quad \text{and} \quad B = \begin{cases} 0.8\alpha_f^{1.28} & \text{if } \alpha_f \leq 0.85 \\ \alpha_f^{2.65} & \text{if } \alpha_f > 0.85 \end{cases} \]

Here, the collision force in Eq. (15) is computed using the soft-sphere model, which decouples its normal and tangential components. The normal force on a particle involved in a collision is given by

\[ F_{\text{col}}^n = (K\delta + \gamma(\mathbf{u}_{12}\mathbf{e}))\mathbf{e} \]  

(20)

In Eq. (20), \( \delta \) is the overlap between the particle pair involved in the collision as illustrated in Figure 4.1 and \( \gamma \) is the damping coefficient, a function of the particle coefficient of restitution \( \eta \); \( \mathbf{e} \) is the unit vector in the direction of \( \mathbf{u}_{12} \). Previous research by Link has demonstrated that for large values of \( K \), the results of the soft-sphere model are identical to those obtained using a hard-sphere model [48]. The tangential collision force is a fraction \( \mu \) of the normal force with \( \mu \) as a function of the relative tangential velocity \( v_r \), given as

\[ \mu(v_r) = \begin{cases} \mu_{\text{stick}} + (\mu_{\text{stick}} - \mu_{\text{glide}})(v_r/v_{\text{glide}} - 2)(v_r/v_{\text{glide}}) & \text{if } v_r < v_{\text{glide}} \\ \mu_{\text{glide}} & \text{if } v_r \geq v_{\text{glide}} \end{cases} \]  

(21)

Figure 4.1. Schematic of particle collision model for DEM [35]
The chemical reactions in the flow require additional equations to compute the local mass fraction of each species $Y_j$ in the computational cell. The species conservation equation is given by

$$\frac{\partial}{\partial t}(\rho Y_j) + \nabla \cdot (\rho u Y_j) = -\nabla \cdot J_j + R_j + S_j$$  \hspace{1cm} (22)

where $J_j$ is the diffusion flux of the species due to concentration gradients in the flow field, $R_j$ is the net rate of production of the species due to chemical reactions, and $S_j$ is the rate of creation of the species from devolatilization.

The oxygen carrier reduction reaction is assumed to occur at the particle surface and is modeled using the multiple surface reactions model (ANSYS, 2012b). To understand this model, consider a reacting particle as shown in Figure 4.2.

![Figure 4.2. Reacting particle in the multiple surface reactions model](image)

For a reaction of order 1, the depletion rate of particle species $j$ in kg/s is given by

$$\bar{R}_j = A_p \eta_r Y_j p_n \frac{R_{kin}D_0}{D_0 + R_{kin}}$$  \hspace{1cm} (23)
where $A_p$ is the spherical surface area of the particle, $\eta_r$ is the effectiveness factor, $Y_j$ is the mass fraction of the species $j$, $p_n$ is the bulk partial pressure of the reacting fluid species $n$, and $D_0$ is the diffusion rate coefficient for the reaction defined as

$$D_0 = C_1 \left[ \frac{(T_p + T_\infty)}{2} \right]^{0.75}$$

where $C_1$ is the diffusion rate coefficient obtained empirically. The final term in the Eq. (24) for the depletion rate, $R_{kin}$, is obtained from the Arrhenius rate equation for the reaction considered. The net depletion of particle mass $dm_p/dt$ due to the reduction of the metal oxide provides the source term $\dot{m}_{s,g}$ used in the continuity equation for the fluid phase.

Heat transfer to the particle is governed by the equation for particle heat balance, which can be written as

$$m_p c_p \frac{dT_p}{dt} = h A_p (T_\infty - T_p) - f_h \frac{dm_p}{dt} H_{reac}$$

where $c_p$ is the particle heat capacity, $h$ is the convective heat transfer coefficient, $H_{reac}$ is the heat released by the reaction and $f_h$ is the fraction of the energy produced that is captured by the particle; the remaining portion $(1 - f_h)$ is applied as the heat source in the energy equation. Values of $f_h$ can range from 1.0 for incomplete combustion where all the heat is retained by the particle (e.g., char combustion to form CO) to zero implying all the heat is released to the continuous phase [35]. Since the reduction of the metal oxide is considered to be a complete reaction, the default value of zero is used in the simulations in this section.
4.2 Simulation of Spouted Fluidized Bed using Fe$_2$O$_3$ as Bed Material

Link [48] identified the different solid-gas flow regimes in multiphase flow in a fluidized bed with relatively large diameter particles of glass beads with a density of 2,500 kg/m$^3$. In recent years, Lagrangian multiphase simulations have captured the particle dynamics in spouted fluidized beds for particle densities ranging from 1,500 to 2,500 kg/m$^3$ [29, 30, 49]. In this section, chemical reactions are incorporated into the CFD-DEM model of a spouted fluidized bed reactor of Zhang et al. [49] while employing particles of Fe$_2$O$_3$ with a density of 5,240 kg/m$^3$ as the bed material. As such, the results of this simulation will also provide an understanding of the effect of material density on the performance of the spouted fluidized bed reactor.

The geometry and computational model of the CD-CLC reactor are shown in Figure 4.3. The geometry is derived from the pseudo-2D Plexiglas test rig used in the TU-Darmstadt cold-flow experiment [30] with the chute structure added to improve particle circulation based on the work of Zhang et al. [49]. The mesh is generated such that the solution is stable when using second-order numerical schemes and minimal under-relaxation to achieve faster convergence at each time step. The particle diameter is kept constant at 2.5 mm; any smaller sized of particles makes the total number of particles required to maintain a reasonable bed height prohibitively large for individual particle tracking in the computation. However, the total particle load in the bed is approximately doubled compared to the work of Zhang et al. [49] to partly offset the tighter packing associated with the heavier particles. Additional particles are also deposited in the down-comer and loop-seal to ensure adequacy of particles for recirculation; a total of 87,320 particles are employed in the entire system.
According to Link [48], the flow regime inside the fluidized bed depends on the ratio of the spout jet velocity and the background velocity to the minimum fluidization velocity of the particles; in order to achieve a spouted fluidized bed, these velocities must be increased in the current simulation compared to the velocities used by Zhang et al. [49] with 2,500 kg/m³ density particles to compensate for the higher minimum fluidization velocity of the heavier Fe₂O₃ particles. The velocities must also be increased to counteract the reduced momentum transfer from the gas phase to the solid phase in the presence of chemical reactions. Thus, the central jet velocity is increased to 50 m/s and the background velocity is increased to 2 m/s.

The flow injection in the fuel reactor consists of 10% CH₄ and 10% H₂O by mass fraction. The reaction kinetics for the reduction of Fe₂O₃ to Fe₃O₄ by the CH₄ are based on the experimental work of Son and Kim [50]. The remaining 80% of the flow injection is inert nitrogen. The absence of CO₂ in the fluidization gas is maintained so that the mass fraction of the CO₂ generated by the reaction between Fe₂O₃ and CH₄ can be tracked without it being overshadowed by the injected mass fraction of CO₂. Similarly, the aeration gas in the down-comer and the loop-seal comprises
solely of N\textsubscript{2} so that the recirculation of particles from the loop-seal to the fuel reactor can be easily identified; the particles that originate in the loop-seal will have a smaller mass fraction of Fe\textsubscript{3}O\textsubscript{4} since they were initially exposed to inert flow. All other parameters in the simulation are kept unchanged from the cold-flow simulation work of Zhang et al. [49]. The key numerical parameters in the present simulation are summarized in Table 4.1.

| Table 4.1. Key modeling parameters for reacting flow simulation in the CLC fuel reactor |
|---------------------------------|---------------------------------|
| Average diameter of particles   | 2.5 mm                          |
| Average density of particles    | 5240 kg/m\textsuperscript{3}    |
| Mass load of particle in bed    | ~1.3 kg                         |
| Primary phase                   | Gaseous mixture of CH\textsubscript{4}, H\textsubscript{2}O, CO\textsubscript{2}, N\textsubscript{2} |
| Secondary phase                 | Solid mixture of Fe\textsubscript{2}O\textsubscript{3}, Fe\textsubscript{3}O\textsubscript{4} |
| Outlet boundary condition       | Pressure outlet with P\textsubscript{out,gage} = 0 Pa |
| Inlet boundary condition        | Velocity inlet with central jet velocity of 50 m/s, background flow velocity of 2 m/s |
| Drag law                        | Syamlal-O’Brien                  |
| Particle collision law          | Spring-dashpot                   |
| Spring constant                 | 410 kN/m                         |
| Coefficient of restitution      | 0.97                             |
| Friction coefficient            | 0.5                              |
| Numerical schemes               | Phase coupled SIMPLE, 2nd-order upwind for momentum equation, QUICK for volume fraction, 2nd-order upwind for energy, 2nd-order upwind for species; 2nd-order implicit in time |
| Time step size                  | Particle: 1×10\textsuperscript{-5} s, Fluid: 1×10\textsuperscript{-4} s |
The simulation is carried out on a Dell workstation using a six-core Intel Xenon CPU. Each run requires about 24 hours of CPU time per 200 ms of simulation time. The particle distributions and velocities are inspected at 40 ms intervals and presented in Figure 4.4.

The particle tracks in Figure 4.4 show that a prominent bubble forms almost immediately as a result of the initial pressure build-up in the fuel reactor. However, owing to the increased particle mass, the velocity of the particles is not sufficient to carry the particles all the way to the top of the reactor and into the cyclone. The bubble bursts around 520 ms as evidenced by the velocity of the particles at the top of the bubble reaching zero and the particles start to fall back into the fuel reactor bed. Moreover, despite the increased total number, the particles initially deposited into the down-comer and loop-seal become tightly packed at the bottom because of the increased particle mass. As a result, there are not enough particles available to recirculate back into the fuel reactor.
Figure 4.4 indicates that the performance of the spouted fluidized bed has a strong dependence on the bed mass, which in turn, depends on the density of the bed material. With the density of Fe$_2$O$_3$ of 5240 kg/m$^3$, the bubble formed in the fuel reactor does not have sufficient energy to reach the top of the reactor. Increasing the central jet velocity further may prevent this issue, but it leads to the formation of a straight pathway through the dense bed region once the initial pressure build-up is lost, which prevents the critical pressure build-up required for subsequent bubbles. Such a pathway is already visible to a certain extent in Figure 4.4 from around 680 ms onwards. Another alternative is to reduce the height of the reactor such that the reduced momentum transferred to the particles is still sufficient for the particles to reach the top of the reactor and into the cyclone. However, this also reduces the residence time of the fuel and affects the fuel conversion rate, particularly using solid coal fuel, which has to go through a slow gasification process before it can react with the metal oxide. Since the particles tracks in Figure 4.4 clearly demonstrate that the spouted fluidized bed setup employed in this section is poor choice, any additional results such as the discussion on the chemical reactions is deferred to the next section in the interest of conciseness.

### 4.3 Simulation of Spouted Fluidized Bed using Fe$_2$O$_3$

**Supported on MgAl$_2$O$_4$ as Bed Material**

In order to address the underlying problem of poor fluidization associated with the high density Fe$_2$O$_3$ particles, it is proposed to use an oxygen carrier consisting of an active part based on Fe combined with an inert support material. The support material is porous and serves to increase the reaction kinetics by providing a larger surface area for reaction as well as to reduce the overall density of the particle. Suitable candidates are MgAl$_2$O$_4$, SiO$_2$, among others [51]. Of these, the
work of Johansson et al. [52] showed that the oxygen carrier consisting of 60% Fe₂O₃ by mass on 40% MgAl₂O₄ support sintered at 1100°C, designated F60AM1100, displayed excellent reactivity and sufficient hardness and its apparent density of 2,225 kg/m³ makes it an ideal choice for fluidized bed operation. In this section, the performance of a spouted fluidized bed reactor with chemically reactions is investigated using the F60AM1100 oxygen carrier as the bed material.

Since MgAl₂O₄ is inert, the only reaction that takes place on the particle surface is the reduction of Fe₂O₃ to Fe₃O₄ by the injected CH₄ fuel. The stoichiometric reaction is given by

\[
12\text{Fe}_2\text{O}_3 \ (s) + \text{CH}_4 \rightarrow 8\text{Fe}_3\text{O}_4 \ (s) + \text{CO}_2 + 2\text{H}_2\text{O}
\]  

(26)

Because of the lower density of F60AM1100, the fluidization velocity in the fuel reactor is reduced to 40 m/s and the total particle load is around 0.7 kg. All other parameters in this simulation remain unchanged from Table 4.1. The computational cost for this setup is similar to that described for the previous section. Figure 4.5 shows the particle distributions and velocities at 80 ms intervals for the first 1600 ms of flow injection.
Figure 4.5 shows that a prominent gas bubble forms in the CD-CLC reactor from 0 to around 400 ms. The leading front of the spout reaches the top of the fuel reactor around 400 ms and a large number of particles are deposited into the cyclone through the connecting duct. Between around 480 ms and 880 ms, the pressure build-up in the fuel reactor vanishes and the remaining particles in the fuel reactor fall back into the fluidized bed while the particles in the cyclone fall into the down-comer. Some recirculation of the particles from the loop-seal back into the fuel reactor is also evident. Once the particles start to settle back into the fluidized bed, aided by the recirculation of particles from the loop-seal, the pressure build-up due to the jet injection is partially restored and subsequent gas bubbles are formed around 960 ms and 1440 ms. However, in these cases, the kinetic energy transferred to the particles is insufficient to carry them to the top of the reactor and the bubbles collapse prematurely. This can be explained by the bypass pathway formed at the same
time by the high velocity jet in the absence of the initial packed bed, which allows the energy in
the jet to bypass the dense bed region and prevents the critical pressure build-up in the fuel reactor.
Since the gas bubble formation and the particle recirculation are both driven by the pressure at
various locations in the system, the static pressure readings at pressure taps P1 through P5 are
investigated to better understand the behavior observed from the particle tracks in Figure 4.5; the
pressure tap data is presented in Figure 4.6.

The subplot at 400 ms shows a large pressure build-up of around 1900 Pa at P1 (base of the
reactor). At 800 ms, when the bubble has collapsed, the initial pressure build-up is lost as the
pressure at P1 drops to around 1250 Pa. Subsequently, at 1200 ms and 1600 ms, the pressure at P1
increases slightly to around 1400 Pa and 1350 Pa respectively. This build-up of pressure is in line

Figure 4.6. Static pressure at pressure taps P1–P5 in the CD-CLC system of Figure 4.3 at t=400 ms (top left), 800 ms
(top right), 1200 ms (bottom left), and 1600 ms (bottom right) in reacting flow
with the observation of the second and third gas bubbles formations; the slight increase compared
to the initial bubble also explains why the subsequent bubbles did not carry sufficient kinetic
energy to reach the top of the reactor. From Figure 4.6, it can also be noted that there is a consistent
positive pressure differential between taps P4 (base of the loop-seal) and P1 of around 100 Pa,
which corroborates the continuous recirculation of particles from the loop-seal back into the fuel
reactor observed from the particle tracks.

Based on the diameter and density, the F60AM1000 particles used as the oxygen carrier can be
classified as Group D particles according to Geldart’s powder classification [17]. In the absence
of experimental results of spouted fluidized bed operation of reacting flow with Group D particles,
the successful incorporation of chemical reactions into the multiphase flow simulation is judged by
inspecting the formation of $\text{Fe}_3\text{O}_4$ and $\text{CO}_2$ as a result of the reaction described in Eq. (26). These
results are presented in Figure 4.7 and Figure 4.8 respectively.

Figure 4.7. Particle tracks colored by mass fraction of $\text{Fe}_3\text{O}_4$ relative to original mass of $\text{Fe}_2\text{O}_3$
From Figure 4.7, it can be seen that the mass fraction of Fe₃O₄ increases with time for the particles inside the fuel reactor as the simulation advances, as expected. The continuous particle recirculation is also evident from the consistent presence of a small number of particles in the fuel reactor with a lower mass fraction of Fe₃O₄ since these particles originated in the loop-seal region where the flow is inert. According to Figure 4.8, the mass fraction of CO₂ rises quickly in the first 640 ms of simulation, after which it drops slightly before spiking again around 1120 ms. Such fluctuations are due to the inherently unsteady nature of the solid-gas mixing in the fuel reactor and similar results are reported in the literature for fluidized bed reactors [53] although none of them have employed the Group D particles used in this study. Overall, the observations from Figure 4.7 and Figure 4.8 indicate successful incorporation of chemical reactions into the CFD/DEM model for the CLC reactor configuration developed by Zhang et al. [49].

Comparing Figure 4.5 with Figure 4.4 in section 4.2, it is clear that changing the bed material in the fuel reactor from Fe₂O₃ to F60AM1100 consisting of 60% Fe₂O₃ and 40% MgAl₂O₄ significantly improved the fluidization performance of the reactor. In the current simulation, the inadequacy of the central jet to impart sufficient momentum to the particles for them to reach the
top of the reactor was rectified and continuous particle recirculation was observed through the loop-seal. However, the formation of the bypass pathway through the bed after the first bubble collapses remains a concern and hinders the pressure build-up required for subsequent bubbles to reach the top of the reactor. Since F60AM1100 is already among the lightest Fe-based oxygen carriers studied by Johansson et al. [52] and lighter alternatives likely to be expensive, one alternate way to mitigate this problem is to use a cyclic flow injection whereby the jet is turned off intermittently to allow the bed particles to re-settle down into the original packed bed configuration, which resets the fluidization behavior to the initial bubble formation stage once the jet is turned back on. Cyclic injections have already been used in laboratory scale CLC experiments such as in the work of Son and Kim [50] to switch between N\textsubscript{2} and CH\textsubscript{4} in lieu of separate fuel and air reactors and their operational feasibility in an industrial setting can be readily studied.

4.4 Simulation of Spouted Fluidized Bed with Pseudo Coal Injection

The spouted fluidized bed was proposed as a viable configuration for CLC with direct coal injection. The addition of coal particles into the fluidized bed system investigated in sections 4.2 and 4.3 would convert the system into a binary particle bed and the collisions between the oxygen carrier and coal would require special attention. Such a system is investigated later in Chapter 6. In this section, only the reaction mechanisms for solid coal are investigated with a pseudo-coal injection that represents the gaseous products of coal after the devolatilization and gasification steps occur. The pseudo-2D reactor in the experiment at TU-Darmstadt used in the reacting flow simulations in the previous sections is considered again with the geometry and mesh shown in Figure 4.3.
The work of Merrick [54] provides a methodology for predicting the compositions of char, tar, and the volatile species based on the physical and chemical properties of the coal. The proximate and ultimate analysis of the South African coal considered in the pseudo-coal simulation is given in Table 4.2 [55].

Table 4.2. Physical and chemical properties of South African coal

<table>
<thead>
<tr>
<th></th>
<th>Proximate Analysis (wt. %)</th>
<th>Ultimate Analysis (wt. %)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Moisture</td>
<td>Volatile matter</td>
</tr>
<tr>
<td>South African coal</td>
<td>8.3</td>
<td>21.6</td>
</tr>
<tr>
<td>Dry ash-free basis</td>
<td>–</td>
<td>28.5</td>
</tr>
</tbody>
</table>

The approach suggested by Merrick [54] to predict the final yields of the volatile matter species is to construct a set of simultaneous linear equations written as $A_{ij}m_j = b_i$ where $m_j$ is the vector of unknowns representing the yields of char (coke), CH$_4$, C$_2$H$_6$, CO, CO$_2$, tar, H$_2$, H$_2$O, NH$_3$, and H$_2$S as mass fractions of the daf coal, and $A_{ij}$ and $b_j$ are matrix and vector of constants respectively. The list of species considered by Merrick [54] is not exhaustive; additional volatile matter species could be modeled if suitable data were available. The first five equations represent element balances on carbon, hydrogen, oxygen, nitrogen, and sulfur respectively. The corresponding values in $A_{ij}$ (for $i = 1, \ldots, 5$ and $j = 1, \ldots, 10$) represent the analyses of the volatile matter species expressed as the mass fractions of the respective species in $b_j$ obtained from the ultimate analysis of the daf coal. The char and tar species are assumed to have a specific composition in terms of the constituent elements given in Table 4.3.
The sixth equation describes the yield of char as a function of the total volatile matter release obtained from the proximate analysis. Merrick [54] obtained a correlation for predicting the volatiles released by coal as

\[ V = p - 0.36p^2 \]  \hspace{1cm} (27)

where \( p \) is the volatile matter from the proximate analysis in the daf coal. For the South African coal considered in the current study with \( p = 0.285 \), the total volatiles released is \( V = 0.256 \). The remaining four equations provide the yields of the remaining volatile species in terms of the ultimate analysis based on approximate evolutions of the hydrogen and oxygen species in the coal in the final volatile matter yield. Merrick [54] found that the yield of \( \text{CH}_4 \) and \( \text{C}_2\text{H}_6 \) accounted for 32.7% and 4.4% of the hydrogen in the coal respectively, and that 18.5% and 11.0% of the oxygen in the coal evolved in the \( \text{CO} \) and \( \text{CO}_2 \) species. Based on these assumptions, the final set of simultaneous equations to obtain the total volatile matter yields can be written as

\[
\begin{bmatrix}
0.98 & 0.75 & 0.8 & 0.4286 & 0.2727 & 0.85 & 0 & 0 & 0 & 0 \\
0.002 & 0.25 & 0.2 & 0 & 0 & 0.082 & 1 & 0.1111 & 0.1765 & 0.0588 \\
0.002 & 0 & 0 & 0.5714 & 0.7273 & 0.049 & 0 & 0.8889 & 0 & 0 \\
0.01 & 0 & 0 & 0 & 0 & 0.009 & 0 & 0 & 0.8235 & 0 \\
0.006 & 0 & 0 & 0 & 0 & 0.01 & 0 & 0 & 0 & 0.9412 \\
1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 \\
\end{bmatrix}
\begin{bmatrix}
\text{Char} \\
\text{CH}_4 \\
\text{C}_2\text{H}_6 \\
\text{CO} \\
\text{CO}_2 \\
\text{Tar} \\
\text{H}_2 \\
\text{H}_2\text{O} \\
\text{NH}_3 \\
\text{H}_2\text{S} \\
\end{bmatrix}
= \begin{bmatrix}
\text{C} \\
\text{H} \\
\text{O} \\
\text{N} \\
\text{S} \\
1-\text{V} \\
1.31\text{H} \\
0.22\text{H} \\
0.32\text{O} \\
0.15\text{O} \\
\end{bmatrix}
\]

\hspace{1cm} (28)

The final yields of the volatile matter and char can be determined by calculating \( \mathbf{m}_j = A_{ij}^{1} \mathbf{b}_i \) and can be written in terms of the mass fractions as

---

45
1 kg daf coal → 0.744 char + 0.0277 tar + 0.0605 CH₄ + 0.0102 C₂H₆ + 0.0325 CO + 0.0152 CO₂ + 0.0140 H₂ + 0.0777 H₂O + 0.0131 NH₃ + 0.00477 H₂S

(29)

Considering the base South African coal with ash and moisture,

1 kg coal → 0.758 daf coal + 0.159 ash + 0.083 moisture → 0.744 char + 0.0277 tar + 0.0605 CH₄ + 0.0102 C₂H₆ + 0.0325 CO + 0.0152 CO₂ + 0.0140 H₂ + 0.0777 H₂O + 0.0131 NH₃ + 0.00477 H₂S

(30)

The mass balance in Eq. (30) can be converted into a mole balance by considering the molecular weight of each species based on its constituent elements. The molecular weight of ash is assumed to be 100 kg/kmol. The final yield from primary devolatilization of 1 kmol of South African coal is given by

1 kmol coal → 0.684 char + 0.0221 tar + 0.0426 CH₄ + 0.00381 C₂H₆ + 0.00390 CO + 0.0131 NH₃ + 0.00477 H₂S + 0.0236 ash + 0.0685 moisture

(31)

Before the gaseous injection prescribing the devolatilization products can be implemented into the reacting flow simulation, the fate of the solid char and tar species in Eq. (31) must be resolved. This is done by considering the gasification of char by steam given by the stoichiometric relation

Char → 2 CO + 0.842 H₂ + 0.00153 H₂O + 0.00438 N₂ + 0.00230 H₂S

(32)

and the secondary devolatilization of tar determined by Bradley et al. [56] assuming a molar ratio of CH₄ to H₂ of 0.5 similar to the primary devolatilization step written as

Tar → 0.805 Cₙsoot + 0.142 CH₄ + 0.0432 CO + 0.285 H₂ + 0.00907 HCN + 0.00441 H₂S

(33)

Hence, the final molar balance after the complete devolatilization and gasification steps is given by
1 kmol coal → 0.0105 C_soot + 0.0269 CH₄ + 0.00224 C₂H₆ + 0.814 CO + 0.00230 CO₂ + 0.0536 H₂ + 0.0292 H₂O + 0.00510 NH₃ + 0.00191 H₂S + 0.0139 ash + 0.0403 moisture

The product species in Eq. (34) are used to specify the pseudo-coal injection for the reacting flow simulation of coal with the Fe-based oxygen carrier F60AM1100 consisting of 60% Fe₂O₃ by mass on inert MgAl₂O₄ support. The injection mole flow rate is set at 0.119 kmol/s of coal to represent a mass injection rate of 1.2 kg/s with a central jet velocity of 30 m/s. The mole fractions of the reacting species, namely CH₄, C₂H₆, CO, and H₂, are kept intact. The remaining species in Eq. (34) are collected into a single inert injection of N₂ for simplicity.

Since the devolatilization and gasification of the coal are already accounted for by the pseudo-coal injection, the only reaction mechanisms implemented in the simulation are for the reduction of Fe₂O₃ by CH₄, C₂H₆, CO, and H₂ to form Fe₃O₄, given as

12 Fe₂O₃ + CH₄ → 8 Fe₃O₄ + 2 H₂O + CO₂
(35)

21 Fe₂O₃ + C₂H₆ → 14 Fe₃O₄ + 3 H₂O + 2 CO₂
(36)

3 Fe₂O₃ + CO → 2 Fe₃O₄ + CO₂
(37)

3 Fe₂O₃ + H₂ → 2 Fe₃O₄ + H₂O
(38)

The reactions are incorporated into the CFD-DEM simulation using the particle surface reactions model outlined in section 4.1. The reaction rates for the metal oxide reduction reactions follow the Arrhenius rate equation \( k = k_0 \exp(-E_a/RT) \) where \( k \) is the reaction rate, \( k_0 \) is the pre-exponent factor, \( E_a \) is the activation energy, \( R \) is the universal gas constant (= 8.314 J/K/mol) and \( T \) is the temperature of the fluid phase. The values of \( k_0 \) and \( E_a \) are obtained from Mahalatkar et al. [22] and are summarized in Table 4.4. It should be noted that the reaction rate for the reduction with C₂H₆ is assumed to be the same as that with CH₄ because of the lack of experimental data.
Table 4.4. Pre-exponent factor and activation energy for the metal oxide reduction reactions

<table>
<thead>
<tr>
<th>Reducing agent</th>
<th>$k_0$ (1/s)</th>
<th>$E_a$ (kJ/mol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH$_4$</td>
<td>$5.33 \times 10^{-4}$</td>
<td>24.0</td>
</tr>
<tr>
<td>CO</td>
<td>$6.20 \times 10^{-4}$</td>
<td>20.0</td>
</tr>
<tr>
<td>H$_2$</td>
<td>$2.30 \times 10^{-3}$</td>
<td>24.0</td>
</tr>
</tbody>
</table>

The particle tracks colored by velocity magnitude are shown in Figure 4.9. An initial bubble forms that carries the bulk of the particles to the top of the fuel reactor and into the cyclone around 400 ms while the remaining particles fall back into the bed. Once the particles settle back down in the bed, a second bubble starts to develop around 900 ms. However, the formation of a bypass pathway is again evident as in Figure 4.5 and gauging by the particle velocities, it is expected that the second bubble will not reach the top of the reactor.

Figure 4.9. Particle tracks colored by velocity magnitude of F60AM1100 particles in reacting flow with a gaseous injection representing the products of devolatilization and gasification of coal
Since the flow conditions and physical properties of the solid and gas phases are unchanged from the simulation in section 4.2, the fluidization behavior is similar as expected. The results of the reaction mechanisms with coal provide greater insight. One measure of the progress of the reaction is the change in the solids mass load in the fuel reactor as shown in Figure 4.10. As the active part of the oxygen carrier, Fe$_2$O$_3$ is reduced to Fe$_3$O$_4$, the total solid mass reduces. The rate of change suggests that the reaction starts out fast and starts to decrease as time advances as the surface area of Fe$_2$O$_3$ available for reaction reduces due to the formation of Fe$_3$O$_4$. A similar trend was observed in the work of Mahalatkar et al. [22] using the Eulerian multi-fluid approach to model the CLC reactor of Leion et al. [55] using solid coal.

![Figure 4.10. Change in solids mass load in the CD-CLC fuel reactor due to the reduction of F60AM1100 particles by the gaseous products of coal devolatilization and gasification](image)

The conversion fraction of the reacting species, namely CH$_4$, C$_2$H$_6$, H$_2$, and CO, is calculated as a ratio of the outlet molar flow rate from the injection flow rate subtracted from unity and is shown
in Figure 4.11. It should be noted that the initial conversion fraction of 1 for each species is a result of the gases not yet reaching the outlet at the start of the simulation as it takes time to travel through the fuel reactor and does not represent a complete depletion of the species. From around 0.6 s onwards, the conversion fraction represents actual values due to species depletion. Given that the reduction of H\textsubscript{2} as the highest value of the pre-exponent factor \( k_0 \) (see Table 4.4), it makes sense that H\textsubscript{2} has the highest conversion fraction approaching 0.4, followed by CO due to the lower activation energy compared to CH\textsubscript{4}. Experimental data of a spouted fluidized bed system for CLC with reacting flow is not available in the literature. However, the conversion fractions obtained in this simulation are in line with CLC systems using a bubbling or fast fluidized bed with smaller oxygen carrier particles and suggests that the spouted fluidized bed is a viable configuration for CLC once the formation of the bypass pathway can be addressed.

![Figure 4.11. Conversion fraction of the gasifying agents in the CD-CLC fuel reactor due to the reduction of F60AM1100 particles by the gaseous products of coal devolatilization and gasification](image-url)

50
4.5 Summary and Conclusion

In this chapter, coupled CFD/DEM multiphase flow simulations of a chemically reacting flow in a pseudo-2D spouted fluidized bed reactor have been conducted as a follow up to the cold flow simulations performed by Zhang et al. [49]. The initial simulation using Fe$_2$O$_3$ as the bed material demonstrated poor fluidization behavior because of the large particle density of Fe$_2$O$_3$. In particular, the bubble formed did not carry sufficient momentum to reach the top of the reactor and particle recirculation from the loop-seal to the fuel reactor was not evident. Both of these are key factors to successful CD-CLC operation. To address these issues, a lighter bed material consisting of 60 wt. % Fe$_2$O$_3$ supported on MgAl$_2$O$_4$ was considered. The results of the simulation using the lighter material showed significant improvement in both areas and highlighted the importance of considering factors other than cost and energy in oxygen carrier selection for CLC.

Even using the lighter bed material, one deficiency that was noted in the simulation of the spouted fluidized bed in terms of the fluidization performance of the system was that after the particles settle down in the bed after the first bubble, subsequent bubbles formed in the fuel reactor lacked the energy required to reach the top of the reactor and into the cyclone. By investigating the pressures in the system, it was found that the high velocity inlet jet formed a bypass pathway once the initial pressure build-up dissipated that prevented the critical pressure build-up required for subsequent bubbles to develop. Alternative approaches were proposed based on this insight, which can be investigated in future work.

Although experimental data for reacting flow in a spouted fluidized bed fuel reactor for CLC is not available in the literature, the successful incorporation of chemical reactions using the particle surface reactions model into the coupled CFD-DEM multiphase model was verified by examining
the formation of CO$_2$ and Fe$_3$O$_4$ in the fuel reactor as a result of the reduction of the Fe$_2$O$_3$ oxygen carrier by gaseous CH$_4$. Since the spouted bed configuration has been proposed for its advantages when solid coal is used as fuel, the reaction mechanisms with coal were investigated by considering a pseudo-coal injection representing the gaseous products after the devolatilization and gasification of the solid coal and the results show great promise. The successful incorporation of the chemical reactions into the coupled CFD-DEM framework marks an important step towards developing a comprehensive functional model for a complete CD-CLC system with solid coal instead of gaseous fuel, which is essential for the design and future optimization of industrial-scale CD-CLC systems.
Chapter 5

Scaling Methodologies for Spouted Beds

The spouted fluidized bed was proposed by Mathur and Gishler [57] to overcome the limitation of a typical bubbling or fast fluidized bed to handle particles larger than a few hundred micrometers in diameter. Relatively larger particles of the oxygen carrier are beneficial for CD-CLC operation for easier separation of the smaller coal and ash particles from the recirculating oxygen carrier; based on the diameter and density, these particles can be classified as Group D particles according to Geldart’s powder classification [17]. The spouted fluidized bed utilizes a high velocity gas stream to create a local high velocity region at the center of the bed (known as the spout) where the particles and voids (bubbles) move in a structured manner with little radial displacement [58].

The high computational cost of CFD-DEM is the reason behind the scarcity of particle-based models for CLC simulation in the literature to date. Since the computational cost of the DEM approach is driven by the number of collisions between particles, the cost can be prohibitive when large systems are considered. The CFD-DEM approach was employed by Parker [59] to develop a comprehensive model of the circulating reactor system at the National Energy Technology Laboratory with reacting flow for CD-CLC but the complexity of the work was such that it took 81 days to complete the 50 seconds of simulation. Industrial scale reactors can contain several trillions of particles; even a laboratory scale experiment can contain particles numbering in the millions. In contrast, the largest number of particles used in a CFD-DEM simulation is 4.5 million in the work of Tsuji et al. [60], which required months of computing time on 16 CPUs. In order to perform coupled CFD-DEM simulations of CLC reactors in a reasonable time with the laboratory resources
available, it is necessary to develop a robust methodology to scale down the number of particles so that the number of collisions is drastically reduced. Dynamic similarity provides such a methodology. This chapter considers scaled simulations of the spouted fluidized bed experiment of Sutkar et al. [61] employing the dynamic scaling methodologies proposed by Glicksman et al. [62] and Link et al. [63] and characterizes the ability of each scaled model to capture the experimental behavior in contrast to the parcel approach that aims to reduce the number of collisions in a CFD-DEM simulation by replacing clusters of particles with parcels. A new scaling methodology is proposed that improves the accuracy of the scaled model compared to the experiment while simultaneously providing the greatest reduction in the computing cost.

5.1 Description of Experimental Setup

The cold flow experiment of Sutkar et al. [61] consists of a pseudo-2D spouted fluidized bed with draft plates as shown in Figure 5.1. The height \( h \) is the particle entrainment height below the draft plates; it is set at 0.3 m in the experiments. The draft plates address the problem of spout gas bypassing and spout instability observed in Chapter 4 by imposing a restriction on the lateral particle flow between the spout and the annulus. By preventing the particles traveling downwards in the annulus from entering the spout and colliding with the particles traveling upwards, the random fluctuations in the spout are eliminated [64]. In the experiment, a high speed particle image velocimetry (PIV) camera is used to capture the instantaneous particle velocities at various heights. These velocity data are used in this paper to quantitatively compare the performance of the different scaling methodologies.
Sutkar et al. [61] employed 1-mm-diameter particles made of glass with a density of 2,500 kg/m³ or γ-Al₂O₃ particles with a density of 1,040 kg/m³ as the bed material in their experiments, though velocity data is only available for the glass beads. The experiment considers both “spouting with aeration” and “fluidized bed-spouting with aeration” flow regimes corresponding to different ratios of the spout velocity and background flow velocity to the minimum fluidization velocity of the
particles. Here, only the fluidized bed-spouting with aeration condition is considered since it is the flow regime better suited for CD-CLC operation. Since velocity data is not available for the γ-Al$_2$O$_3$ particles, the results also compared with a full-scale numerical study conducted as a follow-up to the experiment by the same group [29].

The simulations in this paper are performed using the commercial CFD simulation package ANSYS Fluent, release version 14.5 [34, 35]. The flow field is computed using the Navier-Stokes equations of fluid motion; the motion of the particles is obtained using Newton’s second law. In order to achieve a coupled CFD-DEM simulation for the multiphase flow, source terms are introduced in the Navier-Stokes momentum equation to capture the solid-gas momentum exchange and in the Newtonian equation of motion to account for forces on the solid particles due to the fluid. The equations of fluid and particle motion are as described in Chapter 3 and Chapter 4 respectively. Since simulations in this chapter are performed within a cold flow framework, the energy and species transport equations are not implemented and the source term for interphase mass transfer in Eq. (2) is identically zero.

5.2 Scaling Methodologies

For a typical CD-CLC system, the computational cost of tracking each individual particle is prohibitive. One simple approach available in ANSYS Fluent for reducing the computing load is to divide the particles into clusters called parcels. The motion of each parcel is determined as a whole by tracking a single representative particle [35]. Parcel collisions are evaluated in the same manner as shown in Figure 4.1 but the mass of the entire parcel is considered, not just that of a single representative particle. The parcel diameter is that of a sphere whose volume is the sum of the volumes of its constituent particles. Hence, specifying a parcel diameter equal to twice the
particle diameter leads to a reduction in the number of objects tracked by the DEM solver by a factor of eight, with an even larger decrease in the number of collisions.

5.2.1 Scaling Methodology of Glicksman et al. [62]

The parcel approach provides a good starting point in reducing the computational cost of the coupled CFD-DEM simulation. More robust scaling methodologies can be derived based on the principles of dynamic similarity. By non-dimensionalizing the governing equations of multiphase flow, Glicksman [65] determined the controlling non-dimensional parameters for gas-solid flows. One equation of note is the Ergun equation, which predicts the gas-solid momentum exchange coefficient considering both viscous and inertial effects.

\[
\frac{\Delta p}{L} = 1.75 \frac{\rho_f (1 - \alpha_f) u_0^2}{d_p \alpha_f^3} + 150 \frac{\mu_f (1 - \alpha_f)^2 u_0}{d_p^2 \alpha_f^3} \tag{39}
\]

The full set of scaling parameters were simplified by Glicksman et al. [62] by isolating the viscous and inertial terms on the right hand side of the Ergun equation. The simplified parameters hold exactly at both low and high values of \( \text{Re}_p \) (i.e., for both viscosity-dominated and inertia-dominated gas-solid flows) and are reasonably accurate for the entire range of conditions where the Ergun equation is applicable for obtaining the interphase momentum exchange. They are particularly applicable to a spouted fluidized bed because the relatively large particle diameters and high fluidization velocity places such a system in the inertia-dominated regime. Glicksman et al. [62] proposed a scaling methodology where the dimensions of the fluidized bed are reduced and the superficial velocity \( u_0 \) is adjusted accordingly to maintain the same Froude number, \( \text{Fr} = \frac{u_0}{\sqrt{gL}} \).

Assuming a geometry scale \( r \) with the subscripts \( \text{ex} \) and \( \text{sc} \) representing the exact and scaled systems respectively, the square of the Froude number can be written as
\[
\frac{u_{0,ex}^2}{gL_{ex}} = \frac{u_{0,sc}^2}{gL_{sc}} \Rightarrow \frac{u_{0,ex}}{u_{0,sc}} = \left(\frac{L_{ex}}{L_{sc}}\right)^{1/2} = r^{1/2}
\] (40)

In turn, the minimum fluidization velocity \( u_{mf} \) is adjusted by reducing the particle diameter to hold \( u_0/u_{mf} \) constant. By definition, the minimum fluidization velocity occurs when the pressure drop in the Ergun equation is equal to the gravitational force of the particle bed, as given by

\[
g\left(\rho_p - \rho_f\right) = 1.75 \frac{\rho_f u_{mf}^2}{d_p \alpha_f^3} + 150 \frac{\mu_f \left(1 - \alpha_f\right) u_{mf}}{d_p^2 \alpha_f^3}
\] (41)

After calculating the minimum fluidization velocity for the original model, the particle scaling factor \( n \) can be found by substituting for the minimum fluidization velocity in Eq. (41) and rearranging to solve the quadratic equation for \( n \) given as

\[
g(\rho_p - \rho_f)n^2 - \frac{1.75 \rho_f \left(1 - \alpha_f\right) \left(r^{1/2} u_{mf,ex}^2\right)}{d_p,ex \alpha_f^3} n - \frac{150 \rho_f \left(1 - \alpha_f\right)^2 \left(r^{1/2} u_{mf,ex}\right)}{d_p,ex^2 \alpha_f^3} = 0
\] (42)

Glicksman et al. [62] demonstrated the utility of their scaling methodology by considering 1/4 and 1/16 scale models of an experimental reactor and matching the solid fraction profiles across the systems. In the context of CFD-DEM, the simplified scaling methodology of Glicksman et al. [62] holds promise because the reduction in particle diameter is smaller than the geometry scale used, allowing for a reduction in the total number of particles required in the system to maintain the same bed height. From Eq. (42), given the properties of glass beads and air and a geometry scale of 1/4, the particle diameter must be scaled by 0.62, resulting in a reduction in the number of particles by a factor of around 15 (= (0.62/0.25)^3).
5.2.2 Scaling Methodology of Link et al. [63]

The scaling approach of Glicksman et al. [62] was derived for scaling experimental fluidized beds to reduce cost; its applicability to CFD-DEM simulations due to the reduction in the number of particles is coincidental. It should be noted that the particle Reynolds number, \( \text{Re}_p \), and the Archimedes number, \( \text{Ar} \) (the ratio of gravitational forces to inertial forces) for the scaled system according to Glicksman et al. [62] are roughly equal to 0.34 and 0.12 times their exact values for the 1/4 and 1/16 scaled models respectively in the limit of very small particles; for the larger particles, the difference is even greater and varies with particle density. The ratio of the superficial velocity to the terminal velocity of the particles \( u_0/u_t \) is also significantly decreased. These are important parameters for accurately capturing the fluidization behavior for a spouted bed that cannot be matched by the Glicksman scaling law.

Link et al. [63] proposed a scaling methodology for CFD-DEM simulation that utilizes the ability to change the physical properties of the materials in a computational model, which is not possible in an experiment, in order to maintain the same \( \text{Re}_p \) and \( \text{Ar} \) in the scaled model. By keeping these two parameters the same, the \( u_{mf} \) is also held constant. Unlike Glicksman, the scaling approach proposed by Link et al. [63] only scales up the particle diameter while retaining the original geometry. Hence, \( \text{Fr} \) remains constant without changing \( u_0 \) and the other scaling parameter used by Glicksman et al. [62], \( u_0/u_{mf} \), is automatically matched. By increasing the particle diameter by a factor of \( n \), the number of particles can be reduced by a factor of \( n^3 \) while maintaining the same particle volume. To maintain the same particle Reynolds number, the dynamic viscosity for the gas phase in the scaled simulation is defined as
\[ \text{Re}_{p,sc} = \text{Re}_{p,ex} \Rightarrow \text{Re}_{f,sc} = \frac{d_{p,sc}}{d_{p,ex}} \frac{\mu_{f,ex}}{\mu_{f,sc}} = n \mu_{f,ex} \quad (43) \]

given that the fluid density does not change. In the same vein, the Archimedes number can be held constant by setting a new particle density for the scaled simulation

\[ \text{Ar} = \frac{g d_p^3}{\mu_f^2} (\rho_p - \rho_f); \quad \text{Ar}_{sc} = \text{Ar}_{ex} \Rightarrow \rho_{p,2} = \frac{(\rho_{p,1} - \rho_{f,1})}{n} + \rho_{f,2} \quad (44) \]

### 5.2.3 New Scaling Methodology based on Terminal Velocity

The scaling law of Link et al. [63] addresses the shortcoming of the simplified Glicksman scaling law [62] by maintaining the same Re\(_p\) and Ar between the original and scaled models as well as by maintaining the same Fr and \(U_0/U_mf\). Unfortunately, the ratio \(U_0/U_t\), a crucial parameter in defining the fluidization behavior in a fluidized bed is reduced to 0.707 of its original value, which can be expected to reduce the spout velocity of the particles in the scaled system. To rectify this, a new scaling methodology is proposed in this paper to keep \(U_0/U_t\) constant in addition to every other non-dimensional parameter used in the other two scaling methodologies.

Hence, the final set of scaling parameters used in the novel approach are Fr and \(U_0/U_mf\) from the simplified scaling law of Glicksman et al. [62], Re\(_p\) and Ar from the scaling law of Link et al. [63] and \(U_0/U_t\). First, the geometry is scaled by a factor \(r\) and \(U_0\) by a factor of \(r^{1/2}\) as in Eq. (40) to keep Fr constant. Similar to Link et al. [63], the physical properties of the materials are changed to match certain non-dimensional parameters independently while not affecting others. In this case, the fluid density is changed depending on the particle scaling factor \(n\) (yet to be determined) to keep Re\(_p\) constant given the change in \(U_0\), therefore
\[ \text{Re}_{p,sc} = \text{Re}_{p,ex} \Rightarrow \rho_{f,sc} = \rho_{f,ex} \frac{d_{p,ex} u_{0,ex}}{d_{p,sc} u_{0,sc}} = \frac{\rho_{f,ex}}{n r^{1/2}} \] (45)

For typical values of Re\(_p\) for a spouted fluidized bed, the terminal velocity is defined as

\[ u_t^2 = 3 \frac{(\rho_p - \rho_f) g d_p}{\rho_f} \] (46)

After calculating the terminal velocity for the original model, one can determine \(n\) by substituting for the scaled fluid density and terminal velocity in Eq. (46) and rearranging to solve the quadratic equation for \(n\) given as

\[ \rho_p n^2 \frac{\rho_{f,ex}}{r^{1/2}} n - \frac{1}{3} \frac{\rho_{f,ex} u_{t,ex}^2 r^{1/2}}{g d_{p,ex}} = 0 \] (47)

Using the particle scaling factor determined by Eq. (47) and the corresponding scaling for the fluid’s density given by Eq. (45), Ar for the scaled model can be found to be equal to its original value and the ratio \(u_0/u_{mf}\) is automatically matched. Of the three scaling methodologies as well as the parcel approach discussed in this section, it is expected that this novel approach will most closely reproduce the fluidization behavior of the original scale experiment in the scaled simulation because it retains the same values for a larger set of non-dimensional parameters.

### 5.3 Computational Setup

For each bed material, glass beads and \(\gamma\)-Al\(_2\)O\(_3\) particles, four different cases are considered that reduce the total number of particles in the bed: the parcel approach, the Glicksman scaling law [62], the Link scaling law [63], and the proposed scaling law based on the terminal velocity. As discussed in section 5.2, the independent variable for the parcel approach and the Link scaling law is the particle scale factor \(n\). A value of \(n = 2\) is chosen for the current simulations while keeping the bed geometry the same. On the other hand, the independent variable for the Glicksman scaling law and
the proposed scaling law is the geometry scale factor $r$; it is set at $r = 0.25$ in line with Glicksman et al. [62] and the particle scale is adjusted as determined by the respective scaling methodology. In all eight cases, $u_{sp}$ and $u_{bg}$ are set at roughly $37.0u_{mf}$ and $1.275u_{mf}$ respectively in accordance with the experiment to model the “fluidized bed-spouting with aeration” flow regime. Although the flow in the spouted fluidized bed setup is turbulent, it is well-established that for gas-solid flows, the effect of turbulence is increasingly negligible compared to the effect of the solids for solid volume fractions above 0.001 [66]. For the simulations in this paper, the effect of turbulence can be ignored without loss of accuracy, in line with the previous work using the CFD-DEM approach [59]. The initial bed height (equal to the bed width) is achieved by releasing a large number of particles into the bed prior to the start of each simulation. The different simulation cases considered in this paper and the simulation parameters are summarized in Table 5.1 and Table 5.2 for glass beads and $\gamma$-Al$_2$O$_3$ particles respectively.

Table 5.1. Summary of scaled test cases with glass beads with adjusted physical properties of gases and solids and comparison of non-dimensional parameters with the exact scale

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Exact scale</th>
<th>Parcel approach</th>
<th>Glicksman scaling law</th>
<th>Link scaling law</th>
<th>Proposed $u_T$-based scaling law</th>
</tr>
</thead>
<tbody>
<tr>
<td>$n$</td>
<td>1</td>
<td>2*</td>
<td>0.572</td>
<td>2</td>
<td>0.707</td>
</tr>
<tr>
<td>$r$</td>
<td>1</td>
<td>1</td>
<td>0.25</td>
<td>1</td>
<td>0.25</td>
</tr>
<tr>
<td>$\rho_p$, kg/m$^3$</td>
<td>2500</td>
<td>2500</td>
<td>2500</td>
<td>1251</td>
<td>2500</td>
</tr>
<tr>
<td>$\rho_f$, kg/m$^3$</td>
<td>1.225</td>
<td>1.225</td>
<td>1.225</td>
<td>1.225</td>
<td>3.463</td>
</tr>
<tr>
<td>$\mu_f$, kg/(m-s)</td>
<td>1.79E-05</td>
<td>1.79E-05</td>
<td>1.79E-05</td>
<td>3.58E-05</td>
<td>1.79E-05</td>
</tr>
<tr>
<td>$u_{mf}$, m/s</td>
<td>0.66</td>
<td>0.66</td>
<td>0.33</td>
<td>0.66</td>
<td>0.33</td>
</tr>
<tr>
<td>Parameter</td>
<td>B1</td>
<td>B2</td>
<td>B3</td>
<td>B4</td>
<td></td>
</tr>
<tr>
<td>---------------------------------</td>
<td>-------------</td>
<td>-------------</td>
<td>-------------</td>
<td>-------------</td>
<td></td>
</tr>
<tr>
<td>$u_t$, m/s</td>
<td>7.75</td>
<td>7.75</td>
<td>4.62</td>
<td>7.75</td>
<td>3.87</td>
</tr>
<tr>
<td>$u_{bg}$, m/s</td>
<td>0.84</td>
<td>0.84</td>
<td>0.42</td>
<td>0.84</td>
<td>0.42</td>
</tr>
<tr>
<td>$u_{sp}$, m/s</td>
<td>24.2</td>
<td>24.2</td>
<td>12.1</td>
<td>24.2</td>
<td>12.1</td>
</tr>
<tr>
<td>$(u_0 / u_{mf})_{sc}$</td>
<td>1</td>
<td>N/A</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>$(u_0 / u_{mf})_{ex}$</td>
<td>N/A</td>
<td>0.286</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>$Re_{p,sc}$</td>
<td>1</td>
<td>N/A</td>
<td>0.187</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>$Ar_{sc}$</td>
<td>1</td>
<td>N/A</td>
<td>0.187</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>$(u_0 / u_t)_{sc}$</td>
<td>1</td>
<td>N/A</td>
<td>0.748</td>
<td>0.707</td>
<td>1</td>
</tr>
<tr>
<td>$(u_0 / u_t)_{ex}$</td>
<td>N/A</td>
<td>0.748</td>
<td>0.707</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>Total # particles</td>
<td>461k</td>
<td>51k</td>
<td>33k</td>
<td>51k</td>
<td>17k</td>
</tr>
<tr>
<td>Particle time step, s</td>
<td>–</td>
<td>2e-5</td>
<td>5e-6</td>
<td>2e-5</td>
<td>5e-6</td>
</tr>
</tbody>
</table>

Table 5.2. Summary of scaled test cases with $\gamma$-Al$_2$O$_3$ particles with adjusted physical properties of gases and solids and comparison of non-dimensional parameters with the exact scale
\begin{table}
\begin{tabular}{lccccc}
\hline
& $u_t$, m/s & 4.99 & 4.99 & 2.88 & 4.99 & 2.49 \\
& $u_{bg}$, m/s & 0.46 & 0.46 & 0.23 & 0.46 & 0.23 \\
& $u_{sp}$, m/s & 13.2 & 13.2 & 0.66 & 13.2 & 6.6 \\
& $\left(\frac{u_0}{u_{mf}}\right)_{sc}$ & 1 & N/A & 1 & 1 & 1 \\
& $\left(\frac{u_0}{u_{mf}}\right)_{ex}$ & & & & & \\
& $\frac{Re_{p,sc}}{Re_{p,ex}}$ & 1 & N/A & 0.310 & 1 & 1 \\
& $\frac{Ar_{sc}}{Ar_{ex}}$ & 1 & N/A & 0.239 & 1 & 1 \\
& $\left(\frac{u_0}{u_t}\right)_{sc}$ & 1 & N/A & 0.711 & 0.707 & 1 \\
& $\left(\frac{u_0}{u_t}\right)_{ex}$ & & & & & \\
\hline
Total # & 460k & 51k & 23k & 51k & 17k \\
particles & & & & & \\
Particle time & – & 2e-5 & 5e-6 & 2e-5 & 5e-6 \\
step, s & & & & & \\
\end{tabular}
\end{table}

The particle scaling factor $n$ for the parcel approach in Table 5.1 and Table 5.2 is the ratio between the parcel diameter and the particle diameter. The parcel approach reduces the computing cost by considering a single representative particle within each parcel to calculate the motion of the parcel. As such, a direct comparison between the non-dimensional numbers does not apply for the parcel approach. It should be noted that for the 1/4 scale models, because of the reduced particle size, the particle time step is reduced from $2e^{-5}$ seconds to $5e^{-6}$ seconds to ensure that the particle collisions are accurately resolved. This increase in computational cost is more than offset by the reduced number of particles to track, and is further offset by the coarser mesh required for the 1/4 scaled models to ensure that the particle volume remains smaller than the minimum cell volume. Given that the goal of the scaling methodology is to reduce the computing cost of the CFD-DEM simulation, the reduction in the total number of particles from roughly 461,000 in the original scale...
system is an important parameter for evaluating the performance of the scaling methodology. The reduction in the number of particles is the cube of the ratio of the particle scale factor $n$ to the geometry scale factor $r$. For each case, a greater reduction can be achieved by increasing the independent scaling factor, but a drastic increase can alter the fluidization behavior of the system (e.g., the particles may no longer fall in the spoutable range). For the simulation cases considered, it can be seen from Table 5.1 and Table 5.2 that the proposed $u_t$-based scaling law offers the largest reduction in the number of particles by up to 95% leading to substantial reductions in the computing time in both cases.

The computational domain used in the simulations is an exact representation of the experimental apparatus of Sutkar et al. [61] shown in Figure 5.1. A quarter-scale domain is used for simulations using the Glicksman and the proposed novel scaling approaches ($r = 0.25$). A structured mesh is generated with 24,000 cells for the original scale model and 4,000 cells for the 1/4 scale model. The difference in number is to ensure that the minimum cell volume remains greater than the particle volume for each simulation, a constraint imposed by the CFD-DEM approach. The meshes used in the simulations conducted in this paper are shown in Figure 5.2. Particle velocity data in the $z$-direction is recorded in the central $xz$-plane at heights of 30 cm and 50 cm (7.5 cm and 12.5 cm in the 1/4 scale models) for comparison with the experiment of Sutkar et al. [61] and with the full-scale simulations conducted by the same group [29]. For simulations with the $\gamma$-Al$_2$O$_3$ particles, only the full-scale simulation results are considered for comparison since detailed experimental data is not available.
5.4 Simulations with Glass Beads

For each simulation with glass beads as the bed material, in cases A1–A4, the particle tracks inside the spouted fluidized bed apparatus are recorded after one second and are qualitatively compared against the experimental data as shown in Figure 5.3. It should be noted that while the particle tracks are instructive for verifying that the general fluidization behavior of the system remains the same in the scaled simulations, an instantaneous snapshot is not a suitable metric for determining the performance of the scaling approaches.
Figure 5.3. Particle tracks after 1 s for scaled simulation cases A1–A4 compared with the experiment [61] and full-scale simulation results [29] (particles tracks are colored by nominal velocity magnitude)

The 1/4-scale simulations in Figure 5.3 (i.e., A2 and A4) are shown at the original scale for easier side-by-side comparison. Each scaled case still shows the “fluidized bed-spouting with aeration” behavior observed in the experiment despite any changes in physical properties required by the respective scaling methodologies. The flow in the annulus is very similar for each case, and matches the experimental behavior. It has been noted by Sutkar et al. [61] that the asymmetry in the annulus seen in the experiment was due to slightly uneven gas distribution and is not of any behavioral significance. The behavior of the particles observed in Figure 5.3 is an improvement
over the full-scale simulation work of Sutkar et al. [29]. In their simulation, the particles inside the draft plates formed clusters that partially blocked the flow leading to an unsteady pulsating flow with varying bed height. None of the present cases A1–A4 indicate the presence of particle clusters inside the draft plates, which is more in line with the experimental results than the full-scale simulation.

The spouting particles in the scaled simulations using the parcel approach and the Glicksman scaling law do not reach the same height as the experiment whereas the Link scaling law slightly exceeds the experimental height; the proposed $u_t$-based scaling law comes closest to the experimental bed height in the instantaneous snapshot. The terminal velocity is the lowest velocity required to lift a particle and carry it out of the fluidized bed. In turn, $u_0/u_t$ is a measure of the excess energy available in the flow to lift the particles to a certain height. Since case A4 is the only scaling methodology that matches the value of $u_0/u_t$ in the experiment, it makes sense that it is the only case to match the bed height as well. It is surprising that the particles using the Link scaling law reach a greater height than the experiment considering the value of $u_0/u_t$ for case A3 is lower than that in the experiment. This can be attributed to the fact that the scaled system in A3 uses larger particles with a lower particle density than the experiment according to the scaling methodology of Link et al. [63] described in section 5.2.2; such particles are inherently more spoutable than the original used in the experiment according to Geldart’s powder classification [17]. It makes sense that the height of the particle tracks in case A2 has the largest discrepancy with the experiment because the scaled model according to the Glicksman scaling law has the most unmatched non-dimensional parameters compared to the experiment, as seen in Table 5.1. Surprisingly, case A1 still attains a reasonable approximation to the bed height despite the relative simplicity of the parcel approach although the spout becomes asymmetric.
The particle tracks in Figure 5.3 confirm that the general behavior of the spouted bed remains unchanged in the scaled simulations considered here. However, in order to accurately characterize the performance of the various scaling methodologies, it is important to consider the particle velocities. The time-averaged particle velocity in the z-direction at two different bed heights of 30 cm and 50 cm is used to quantitatively characterize the accuracy of the different scaling methodologies. Figure 5.4 compares the time-averaged particle z-velocity in the central $xz$-plane at a height of 30 cm for the scaled simulation cases A1–A4 against the experimental results [61] and the results of the full-scale simulation conducted by Sutkar et al. [29].

![Graph](image.png)

Figure 5.4. Time-averaged particle $z$-velocity at $z=30$ cm for scaled simulation cases A1–A4 compared with experiment [61] and full-scale simulation results [29].

The full-scale simulation with 460k particles can predict the peak particle velocity in the spout region but the predicted velocities are lower near the ends of the spout region adjacent to the draft
plates. Both the Glicksman and Link scaling laws can capture the trend in particle velocities in the spout but the exact velocities are lower than the experiment. This discrepancy is expected according to Table 5.1, which shows that \( u_0/u_t \) is around 0.7 for both these scaling methodologies. On the other hand, the proposed \( u_t \)-based scaling law can accurately capture the spout velocity across the entire spout region, though it slightly overshoots the peak velocity at the center of the spout; the proposed law also shows the best results in the annulus. None of the other scaled simulations, or even the full-scale simulation, come close to matching the downward particle velocities in the annulus with the experiment. It should be noted that the parcel approach performed the worst out of all the scaling methodologies considered, which can be explained by the relative simplicity of the approach and the lack of scientific rigor in its formulation.

The time-averaged particle \( z \)-velocity in the central \( xz \)-plane at a height of 50 cm for the scaled simulation cases A1–A4 is compared against the experimental results [61] and the results of the full-scale simulation conducted by Sutkar et al. [29] in Figure 5.5. In this case, there is no clear demarcation between the spout and annulus regions because 50 cm is above the draft plates. The full-scale simulation is a good approximation of the experimental values everywhere except the extreme ends of the bed. On the other hand, the proposed \( u_t \)-based scaling law predicts the experimental values correctly at the extremes as well as in the center, but the particle velocities in the other areas are too low. However, the proposed scaling law still performs the best among the various scaling laws considered; the Glicksman and Link scaling laws under-predict the particle velocities across the entire bed. Once again, the parcel approach performs the worst.
5.5 Simulations with $\gamma$-Al$_2$O$_3$ Particles

The dynamic behavior and fluidization in a gas-solid system for CLC depend on the physical properties of the bed material (particle diameter, density, restitution coefficient, etc.). Therefore, it is important to verify the effectiveness of the proposed $u_t$-based scaling approach for a different bed material. The $\gamma$-Al$_2$O$_3$ particles considered in this section have the same diameter as the glass beads but lower density and restitution coefficient of 1040 kg/m$^3$ and 0.74 respectively compared to 2500 kg/m$^3$ and 0.97 for glass. The time-averaged particle velocity of the $\gamma$-Al$_2$O$_3$ particles in the $z$-direction for simulation cases B1–B4 is recorded at heights of 10 cm and 30 cm respectively and compared against the results from the full-scale simulation of Sutkar et al. [29]. The velocity
profiles using the different scaling methodologies listed in Table 5.2 at 10 cm and 30 cm are presented in Figure 5.6 and Figure 5.7 respectively.

![Graph of particle z-velocity at z=10 cm for scaled simulation cases B1–B4 compared with full-scale simulation results](image)

Figure 5.6. Time-averaged particle z-velocity at z=10 cm for scaled simulation cases B1–B4 compared with full-scale simulation results [29]

It is noted that the full-scale simulations results shown in Figure 5.6 and Figure 5.7 are asymmetric due to the way the boundary conditions were imposed [29]; similar asymmetry could be observed in the particle tracks shown in Figure 5.3 as well as in the velocity profiles in Figure 5.4 and Figure 5.5. As such, only the magnitude of the velocities in the full-scale simulation results should be considered for comparing the scaling methodologies, not the profile shape. At z=10 cm, there is a strong spout in the central region but the particles in the annulus are densely packed. On the other hand, at z=30 cm, the spout velocities are weaker but there is a distinct downwards motion of the particles in the annulus. In both Figure 5.6 and Figure 5.7, the Link scaling law and the proposed
$u_t$-based scaling provide the best match with the full-scale simulation results; the parcel approach and the Glicksman scaling law under-predict the spout velocity. All the scaling laws capture the densely packed bed with no particle velocity in the annulus as shown in Figure 5.6. However, the proposed $u_t$-based scaling law better captures the gradual change in particle $z$-velocities in the annular region in Figure 5.7; the Link scaling law shows a nearly zero velocity in much of the annulus before producing a sharp decrease towards the edges. Once again, the proposed scaling law can be seen to produce the best match with the full-scale simulation results on top of providing the largest reduction in the number of particles.

![Graph showing particle $z$-velocity at $z=30$ cm for scaled simulation cases B1–B4 compared with full-scale simulation results][29]
5.6 Summary and Conclusion

The parcel approach, the simplified scaling law of Glicksman et al. [62], the scaling law of Link et al. [63], and the proposed $u_t$-based scaling law have been used to simulate an experimental spouted fluidized bed with draft plates using the CFD-DEM method. The particle velocity is an important quantity for characterizing the fluidization behavior in a spouted fluidized bed; it is used in this paper to compare the performance of the different scaling approaches. Comparing the particle velocities in the $z$-direction at various heights, it is found that all the scaling methodologies can capture the general trends in the particle velocities at different heights. The proposed $u_t$-based scaling law outperforms the other scaling approaches and provides the best match with the experimental values. This makes sense because the scaled model using the proposed approach maintains the same values as the experiment for all the non-dimensional parameters used to ensure dynamic similarity. The proposed scaling law also provides the largest reduction in the number of particles in the system in all cases, and hence, the largest reduction in computing cost. The establishment of a scaling law that can maintain fidelity with experiment is a crucial step towards the development of CFD-DEM simulations of industrial scale fluidized beds for chemical looping combustion.
Chapter 6

Binary Particle Bed Simulations in a Carbon Stripper

The use of solid coal fuel instead of gaseous fuels in chemical looping combustion introduces additional operational complexities. Unlike gaseous fuel, which is directly combusted by the oxygen carrier, the coal must first undergo a devolatilization process followed by a gasification reaction where the remaining char is reacted by the fluidizing gases consisting of recycled CO$_2$ and/or H$_2$O. The products of devolatilization and gasification are then combusted by the oxygen carrier. A typical CLC setup utilizes a cyclonic separator to isolate the oxygen carrier particles from the flue gases after the fuel reactor and the air reactor before transporting the solids between the reactors to continue to loop. Since the char gasification is a slow process [12], unburnt char particles often remain in the flue stream of the fuel reactor. If these are transported to the air reactor along with the oxygen carrier particles, the carbon capture efficiency of the CD-CLC process would be reduced.

Several approaches have been proposed to prevent char particles from reaching the air reactor. One way is to provide sufficient residence time in the fuel reactor to ensure that the gasification reaction is complete. This can be achieved either by increasing the size of the reactor or by reducing the fluidizing gas velocity, but both options can impede the fluidization behavior of the bed, particularly in a spouted bed configuration as seen in Chapter 4. To avoid the poor fluidization while still maintaining an increased residence time, a multi-staged fuel reaction concept was
recently proposed and investigated [67]. A mass and energy balance study of the multi-staged fuel reaction setup conducted using Aspen Plus demonstrated that complete char conversion can be achieved by using multiple smaller fuel reactors in series such that any unburnt char in the system is burnt in subsequent fuel reactor stages before the solids are transported to the air reactor.

Figure 6.1 shows the differences in size between the particles of pulverized coal and a typical oxygen carrier (ilmenite) used in CD-CLC operation. In fact, one reason of considering the spouted fluidized bed configuration is that it overcomes the limitation of a bubbling or fast fluidized bed to handle particles larger than a few hundred micrometers in diameter. Thus, one way of preventing the leakage of unburnt char into the air reactor is to take advantage of the differences in size and density, and hence the terminal velocity, to separate the lighter char from the heavier oxygen carrier particles. Since char already has a lower density than the oxygen carrier, using pulverized coal particles smaller than or almost of the same size as the oxygen carrier particles should invariably lead to satisfactory separation results. The devolatilization and gasification processes that the coal undergoes further decrease the char particle size, enhancing the separation effect. The device that separates the char particles from the char and oxygen carrier mixture stream exiting the fuel reactor is known as a carbon stripper. The char particles from the carbon stripper can be returned to the fuel reactor to complete the gasification step while the oxygen carrier particles are transported to the air reactor to be regenerated.
Figure 6.1. Size difference between particles of coal and oxygen carrier

By preventing the combustion of the unburnt char in atmospheric air, the carbon stripper also eliminates the formation of pollutants such as CO$_2$ and NO$_x$ in the air reactor, as highlighted by Kramp et al. [68] and Mendiara et al. [69], and is deemed critical for CD-CLC operation despite the increased hydrodynamic complexity associated with implementing the carbon stripper compared to increasing the residence time in the fuel reactor. It is noted that for other fuels such as biomass where the increase in residence time required for the solid fuel is smaller, the direct increased residence time approach may be more competitive. In recent years, carbon strippers operating with fluidizing velocity in the range of 0.15–0.40 m/s have been incorporated into CD-CLC experiments by Markström et al. [70], Ströhle et al. [71], Abad et al. [72], and Sun et al. [73]. The results of these experiments indicated that the fluidization velocity should be increased further to increase the particle separation.

Later, Sun et al. [74] conducted cold-flow studies using a riser-based carbon stripper operating in the fast fluidized bed regime to investigate the effect of gas velocity on the separation ratio. The goal of Sun et al.’s design [74] was to achieve a high separation ratio to minimize the leakage of char particles into the air reactor with a low fluidizing gas velocity to keep operational costs low. However, the specific nature of the multiphase solid-gas flow inside the carbon stripper and how its geometry affects the design targets is not well understood from the experiment. In order to
identify these relationships, a CFD-DEM coupled simulation is developed in this chapter for the carbon stripper consisting of a binary particle bed of coal and oxygen carrier particles and is validated against the experiment of Sun et al. [74]. In future work, there is considerable scope to optimize the geometry of the carbon stripper to enhance the achievement of these design goals that can be addressed by integrating a multi-objective genetic algorithm with the CFD-DEM code.

### 6.1 Description of Experimental Setup

The carbon stripper used in the cold-flow experiment by Sun et al. [74] consisted of a riser, 4 m tall with a diameter of 0.7 m. A schematic of the experimental setup is presented in Figure 6.2. The solids mixture contained 95% ilmenite particles by mass and 5% plastic beads representing the unburnt char particles in the system.

![Figure 6.2. Schematic of riser-based carbon stripper used by Sun et al. [74]](image)
The physical properties of ilmenite and plastic beads are listed in Table 6.1. The riser was fluidized from the bottom by air with the fluidizing velocity \( u_g \) in the range of 1.50–2.75 m/s increasing at 0.25 m/s intervals. \( u_g \) was selected to fall between the terminal velocities \( u_t \) for the ilmenite and plastic beads such that the plastic beads will be carried out of the bed and exit the riser from the top into a tank while the ilmenite particles remain in the bed and collect in the bottom tank. The solids mixture is injected from the side of the riser at a height of \( H_{inlet} = 1 \) m above the bottom collection tank.

Table 6.1. Properties of ilmenite particles and plastic beads used by Sun et al. [74]

<table>
<thead>
<tr>
<th>Particle</th>
<th>( d_p ) (0.5) (μm)</th>
<th>( \rho_p ) (kg/m³)</th>
<th>( u_t ) (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ilmenite</td>
<td>257</td>
<td>4,260</td>
<td>5.65</td>
</tr>
<tr>
<td>Plastic beads</td>
<td>94</td>
<td>960</td>
<td>0.39</td>
</tr>
</tbody>
</table>

The separation ratio \( \lambda \) is defined as the mass of particles collected from top tank to the mass of particles collected from the top and bottom tanks combined, as given by

\[
\lambda = \frac{m_{mix, top}}{m_{mix, top} + m_{mix, btm}}
\]  

The concentration of plastic beads in each mixture sample in the experiment was determined by burning the mixture and measuring the change in weight. The plastic beads completely combusted to form CO\(_2\) and H\(_2\)O while the weight loss of the oxygen carrier was approximately 1% due to the reduction of ilmenite. \( \lambda \) is calculated for each \( u_g \) based on the experimental results and is plotted in Figure 6.3 for a solids mixture feeding rate \( G_{mix,in} \) of 12.2 kg/m\(^2\)-s; the same value of \( G_{mix,in} \) is used in the simulations.
6.2 Computational Setup

The geometry used in the CFD-DEM simulation of the carbon stripper used by Sun et al. [74] uses the exact dimensions of the riser presented in Figure 6.2. Since the solids flow from the feed hopper into the riser is of no consequence to the simulation, the solids inlet is simply modeled as a partial pipe. The top collection tank is also eliminated and the solids flow at the top is measured directly at the riser outlet. The bottom collection tank is modeled as a simple closed boundary in order to ensure the accurate pressure boundary condition at the bottom of the riser; the solids flow into the bottom tank is measured at the surface between the riser and the tank. A structured grid is generated for all elements of the geometry and is shown in Figure 6.4. The total number of cells is 51,884 in order to maintain a minimum cell volume greater than the particle (parcel) volume.
Given the small particle diameters of ilmenite and plastic beads used in the experiment (see Table 6.1), the number of particles in the system is very large. The parcel approach described in section 5.2 is employed to reduce the computational load with a parcel diameter of 0.002 m. Two solids injections are used corresponding to the ilmenite and the plastic beads; the injection mass flow rates are calculated based on $G_{mix,in}$, the riser cross-section $A_r$ ($= \pi D_r^2 / 4$), and the concentration of plastic beads in the solids flow $c_{pb}$, as outlined below.
\begin{equation}
\dot{m}_{mix,in} = \dot{G}_{mix,in}A_r \tag{49}
\end{equation}

\begin{equation}
\dot{m}_{ilm,in} = \frac{\dot{m}_{mix,in}}{1 + c_{pb}} \quad \dot{m}_{pb,in} = \frac{\dot{m}_{mix,in}}{1 + c_{pb}} \tag{50}
\end{equation}

The volumetric flow rates can be determined given the respective densities of the two materials and are used to determine the solids injection velocity.

\begin{equation}
q_{ilm,in} = \frac{\dot{m}_{ilm,in}}{\rho_{ilm}} \quad q_{pb,in} = \frac{\dot{m}_{pb,in}}{\rho_{pb}} \tag{51}
\end{equation}

\begin{equation}
q_{mix,in} = q_{ilm,in} + q_{pb,in} \tag{52}
\end{equation}

\begin{equation}
\dot{u}_{mix,in} = \frac{q_{mix,in}}{A_r} \tag{53}
\end{equation}

As before, the soft sphere model shown in Figure 4.1 is used for all the particle-particle and particle-wall collisions. In order to keep computing time low, the spring stiffness \( k_n \) is set at 5,000 N/m to relax the minimum particle time step requirement. Bokkers [75] demonstrated that the results produced using this value of \( k_n \) are indistinguishable from those using larger values of \( k_n \), which necessitate a smaller particle time step. The coefficient of restitution is set at 0.97. The numerical simulations are conducted using the phase-coupled SIMPLE scheme with 2nd order discretization in space and 1st order in time. The simulation cases modeled and the key modeling parameters are summarized in Table 6.2.

Table 6.2. Key modeling parameters for binary particle bed simulation in the riser-based carbon stripper

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Primary phase</td>
<td>Air</td>
</tr>
<tr>
<td>Discrete phase(s)</td>
<td>Ilmenite; plastic beads</td>
</tr>
<tr>
<td>Parcel diameter</td>
<td>0.2 m</td>
</tr>
<tr>
<td>Gas inlet fluidizing velocities</td>
<td>1.50, 1.75, 2.00, 2.25, 2.75 m/s</td>
</tr>
<tr>
<td>Solids injection velocity</td>
<td>( u_{mix,in} = 0.0034 \text{ m/s} )</td>
</tr>
</tbody>
</table>
Solids injection flow rate \( \dot{m}_{ilm, in} = 0.044 \text{ kg/s}; \dot{m}_{pb, in} = 0.0023 \text{ kg/s} \)

Outlet Pressure outlet at atmospheric pressure

Drag law Gidaspow [37]

Particle collision model Soft-sphere model

Spring constant 5000 N/m

Coefficient of restitution 0.97

Friction coefficient 0.5

Time step size Particle: 5\times10^{-5} \text{ s}; fluid: 5\times10^{-4} \text{ s}

### 6.3 Binary Particle Bed Simulation Results

Each CFD-DEM simulation of the binary particle bed in the riser-based carbon stripper is run for 20 s. The solids flow rate out of the riser outlet and into the bottom collection tank as well as the static pressure in the bed is recorded every 20 time steps (0.01 s). In the experiment of Sun et al. [74], after the initial development of fluidization caused by the solids injection, the pressure differences across sampling ports 1–3 and 8–11 shown in Figure 6.2 stabilized after approximately 10 s. The static pressure at 2 mm above the inlet is used to verify the stable bed in the numerical simulation; the results for the \( u_g = 1.50 \text{ m/s} \) case are given in Figure 6.5. It can be seen that the static pressure in the bed stabilizes after approximately 8 s. The final 10 s of simulation is used as the averaging interval for the solids flow rates in order to calculate the separation ratio \( \lambda \), which is used to quantitatively validate the accuracy of the simulation against the experimental results [74]. To confirm that the averaging interval does not affect the value of \( \lambda \), the simulation with \( u_g = 2.00 \text{ m/s} \) was run for 30 s and the computed difference in \( \lambda \) was miniscule.
The development of solids flow into and out of the riser can be ascertained by examining the number of particles (parcels) of ilmenite and plastic beads held up in the riser after 20 seconds of simulation, as shown in Table 6.3. As $u_g$ increases, the number of ilmenite parcels in the riser increases. This is because the increased gas velocity prevents the ilmenite from settling at the bottom of the riser and flowing into the bottom collection tank. However, this increase starts to diminish around $u_g = 2.50$ m/s and for $u_g = 2.75$ m/s, the number of ilmenite particles in the riser decreases drastically. Although 2.50 m/s is still lower than the terminal velocity of ilmenite, the decreased hold up suggests that at this velocity, the flow has sufficient energy to carry the particles out of the riser. On the other hand, the number of plastic beads in the riser steadily decrease for as the fluidizing velocity increases. This is expected since the $u_g/u_t$ ratio starts out at more than 1 at $u_g = 1.50$ m/s and as it gets larger, the flow is able to carry the plastic beads out with greater ease.

<table>
<thead>
<tr>
<th>Particle</th>
<th>1.50 m/s</th>
<th>1.75 m/s</th>
<th>2.00 m/s</th>
<th>2.25 m/s</th>
<th>2.50 m/s</th>
<th>2.75 m/s</th>
</tr>
</thead>
</table>

Table 6.3. Number of parcels in riser after 20 seconds of simulation for different fluidizing velocities

Figure 6.5. Static pressure at 2 mm for binary particle bed simulation with fluidizing velocity $u_g=1.50$ m/s
For each case, the solids flow out of the riser outlet consists almost entirely of plastic beads with a few ilmenite particles, except at $u_g = 2.75 \text{ m/s}$. On the other hand, the solids flow into the bottom collection tank is solely composed of ilmenite. This is expected given that the fluidizing velocities in each case lies between the terminal velocities of the plastic beads and ilmenite particles such that the fluid can carry the lighter plastic beads out of the bed but not the ilmenite particles. The flow rate of plastic beads out of the top riser outlet for different values of $u_g$ is presented in Figure 6.6. As $u_g$ increases, the plastic beads reach the outlet faster because of a higher induced particle velocity and the overall flow rate increases slightly until it stabilizes at a roughly constant value in each case equal to the injection flow rate of plastic beads; the plastic beads flow rate into the bottom collection tank is nil. The transient fluctuations in the flow rate are due to the highly unsteady flow in the fast fluidization regime associated with the riser.

![Figure 6.6. Plastic beads flow rate out of top of the riser for different fluidizing velocities](image_url)

Figure 6.6. Plastic beads flow rate out of top of the riser for different fluidizing velocities
Similar plots are generated for the flow rate of ilmenite out of the top of the riser and into the bottom collection tank and are shown in Figure 6.7 and Figure 6.8 respectively. As mentioned above, the ilmenite flow rate out of the top outlet is limited to isolated particles up to \( u_g = 2.25 \) m/s. The ilmenite flow rate into the bottom collection tank decreases as \( u_g \) increases. The flow rate plots confirm the solids flow behavior suggested by the parcel hold up numbers in Table 6.3.

![Ilmenite flow rate out of top of the riser for different fluidizing velocities](image)

Figure 6.7. Ilmenite flow rate out of top of the riser for different fluidizing velocities
Figure 6.8. Ilmenite flow rate into bottom collection tank for different fluidizing velocities

The flow rates of the plastic beads out of the top of the riser and the ilmenite into the bottom collection tank shown in Figure 6.6 and Figure 6.8 respectively are used to compute the separation ratio $\lambda$ according to Eq. (48). The values of $\lambda$ for different fluidization velocities are plotted in Figure 6.9. The values of $\lambda$ in Figure 6.9 are in excellent agreement with the experimental values presented in Figure 6.3. Hence, the binary particle bed simulation conducted in this chapter can be considered to be a credible model for the experiment and can be employed to examine additional changes to the geometry and operating conditions and investigate their effect on $\lambda$. 
6.4 Summary and Conclusion

In this chapter, CFD-DEM simulations are conducted of the riser-based carbon stripper developed by Sun et al. [74] consisting of a binary particle bed of plastic beads (corresponding to coal) and ilmenite particles. The static pressure in the bed is used to assess the onset of a stable fluidization regime and the flow rates of the plastic beads and ilmenite out of the riser outlet and into the bottom collection tank respectively are averaged to determine the particle separation ratio. The results show excellent agreement with the experimental data and establish a credible model for a binary particle bed that can be used to optimize the design and operation of such systems in future work.

One point of disagreement between the simulation and the experiment is the absence of plastic beads in the bottom collection tank. In the experiment, a few plastic beads are collected in the bottom tank at lower values of \( u_g \) [74]. However, theoretically, this behavior was unexpected since
even the smallest value of $u_g = 1.50$ m/s is well above the terminal velocity of the plastic beads and should carry the beads out of the riser.
Chapter 7

Future Work

Although agencies such as NETL and companies such as Alstom are investing significant capital into the development of pilot projects in CLC to further develop the technology for deployment on an industrial scale, there has been no concerted research into developing an optimization methodology for the design or operating conditions for the CLC system. The experimental results of Leion et al. [55] and Sun et al. [74] suggest that significant improvements can be obtained in the performance of the CLC system in terms of fuel conversion and particle segregation respectively by changing the operating conditions of the system. The geometry of the CLC reactor plays a significant role in the performance of the system as well. For example, the inclusion of draft plates in the pseudo-2D apparatus considered by Sutkar et al. [61] produced a significantly enhanced fluidization behavior compared to the setup used by Alobaid et al. [30]. Zhang et al. [49] showed that a chute added to the bottom of the fluidized bed eliminated the dead zone of stagnant particles and improved the pressure distribution in the system resulting in an increase in the solids circulation. A comprehensive optimization methodology for the design and operation of CLC is the logical next step to push this technology towards the industrial viability. As the demand of more effective CCS technologies grows, the environmental and economic impact of this work will be immense.

Many of the CFD-DEM simulations conducted in this dissertation focused on the spouted fluidized bed configuration because of its advantages with the relatively larger oxygen carrier particles that are used when considering coal-direct CLC. The reaction kinetics in CD-CLC were investigated
with a pseudo-coal fuel injection in Chapter 4 but a complete CFD-DEM model of a CLC system using solid fuels has not been simulated in the present work. The binary particle bed aspect of a coal-oxygen carrier system was considered in Chapter 6 but by substituting plastic beads for the coal. The inclusion of coal particles in DEM introduces additional complexities such as the changes in diameter and density as the devolatilization and gasification reactions take place; this work should be attempted in the future. The reacting flow studies discussed in Chapter 3 highlight the paucity of reaction rate data for metal oxide reduction reactions other than Fe$_2$O$_3$. There is significant scope for experimental studies to identify accurate reaction kinetics for these reactions to enhance the accuracy of CFD simulations in the future. Reacting flow experiments must also be conducted using the spouted fluidized bed configuration to generate data for validating the simulation results in Chapter 4 that incorporated chemical reactions into the CFD-DEM framework.
References


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