Multi-phase CFD modeling of solid sorbent carbon capture system

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A B S T R A C T
Computational fluid dynamics (CFD) simulations are used to investigate a low temperature post-combustion carbon capture reactor. The CFD models are based on a small scale solid sorbent carbon capture reactor design developed by ADA-ES and Southern Company. The reactor is a fluidized bed design based on a silica-supported amine sorbent. CFD models using both Eulerian–Eulerian and Eulerian–Lagrangian multi-phase modeling methods are developed to investigate the hydrodynamics and adsorption of carbon dioxide in the reactor. Models developed in both FLUENT® and BARRACUDA are presented to explore the strengths and weaknesses of the art CFD codes for modeling multi-phase carbon capture reactors. The results of the simulations show that the FLUENT® Eulerian–Lagrangian simulations (DDPM) are unstable for the given reactor design; while the BARRACUDA Eulerian–Lagrangian model is able to simulate the system given appropriate simplifying assumptions. FLUENT® Eulerian–Eulerian simulations also provide a stable solution for the carbon capture reactor given the appropriate simplifying assumptions.

1. Introduction
Carbon capture and sequestration technologies are being developed as part of the plan for a sustainable energy future. The U.S. Department of Energy has set goals for carbon capture systems at 90% carbon dioxide (CO2) capture with less than a 35% increase in cost of electricity [1]. Carbon capture is the separation of CO2 from the exhaust gas of a power plant for later sequestration. There are several pre- and post-combustion designs being investigated for carbon capture systems. Pre-combustion technologies are applied to gasification systems and are not discussed here. Post-combustion technologies fall into two categories: (1) conventional amines such as methyl-ethyl amine (MEA) or (2) advanced solid sorbent based systems. The advanced solid sorbent systems are further divided into low and high temperature systems. In this work, we discuss the simulated performance of a low temperature supported polyamine based process. Solid sorbent systems offer an advantage over other capture technologies because they can reduce the regeneration energy associated with CO2 capture and reduce parasitic load. Most of the work on sorbents has been restricted to developing the sorbent itself [2], with only recent studies considering the design of the reactor system and integration with the power plant [3,4]. The U.S. Department of Energy’s National Energy Technology Laboratory (NETL) is sponsoring a number of sorbent development efforts; however, significant work remains in defining and optimizing the reactors and process parameters needed for successful implementation of sorbent-based capture systems.

In this paper we will discuss a 1 kWe solid sorbent carbon capture reactor that has been modeled as part of the U.S. Department of Energy’s Carbon Capture Simulation Initiative (CCSI). The work focuses on computational modeling of device-scale multi-phase computational fluid dynamics (CFD) simulations for a given carbon capture reactor design to predict flow properties, outlet compositions, temperatures, and pressures. The detailed outputs of the device-scale model provide valuable insight into the operation of new carbon capture devices and will help in the design and optimization of carbon capture systems. The 1 kWe solid sorbent carbon capture system is based on an experimental setup developed by ADA-ES and Southern Company. This small-scale system was chosen to investigate the capabilities of state of the art CFD modeling codes and to provide insight to ADA-ES on the design and operation of their 1 kWe system. Although the 1 kWe case is much smaller than anticipated full-scale carbon capture systems, it represents the first step in a series of pilot-scale demonstrations utilities perform when evaluating new technologies. It is also the first step toward the development of larger scale CFD models, which are under development as part of CCSI. More importantly, the modeling and technical knowledge gained through the 1 kWe reactor modeling process will be useful in the development of larger pilot-scale systems and corresponding computational models.

The multi-phase reactive transport of the flue gas and the solid sorbent particles through a 1 kWe solid sorbent reactor system is modeled using commercial CFD codes. Two different multi-phase CFD modeling approaches are used to simulate the solid sorbent system; an Eulerian–...
Eulerian model and two different Eulerian–Lagrangian models. Multiple multi-phase modeling methods are considered to investigate the applicability of the models and the various advantages and disadvantages of the available modeling tools and methods. Two different commercial CFD codes are used to model the 1 kWe solid sorbent reactor: ANSYS's FLUENT® and Computational Particle Fluid Dynamic's (CPFD's) BARRACUDA. FLUENT® offers multiple modeling tools for multi-phase flow including Eulerian–Eulerian and Eulerian–Lagrangian modules; while, BARRACUDA uses a particle based Eulerian–Lagrangian modeling method. The results of three different CFD models (FLUENT® Eulerian–Eulerian, FLUENT® Eulerian–Lagrangian and BARRACUDA Eulerian–Lagrangian) of the solid sorbent system are presented in this paper. The advantages and disadvantages of each model are discussed along with areas for further improvement and development of CFD codes.

In this paper, we will discuss the details of the solid sorbent carbon capture system being modeled; the multi-physics included in the CFD models; the CFD modeling tools and simulation domain; and the Eulerian–Eulerian and Eulerian–Lagrangian CFD models including results and comparisons between the different CFD codes. The goals of this paper are to better understand the multi-physics and the operation of the solid sorbent system, to investigate the accuracy of modeling the system using current state-of-the-art CFD modeling tools, and to explore the benefits and weaknesses of using different multi-phase modeling methods for simulating solid sorbent carbon capture systems.

2. Solid sorbent carbon capture system

The 1 kWe carbon capture system considered in this paper is a fluidized loop consisting of three components: the riser, internally circulating fluidized bed (ICFB), and regenerator, per the schematic illustration in Fig. 1. CO₂-containing flue gas enters the carbon capture system at the riser, and solid sorbent particles are introduced in the riser upstream of the flue gas inlet. The adsorption of CO₂ occurs in the riser as the particles and gas flow up the riser to the ICFB, where CO₂ adsorption continues. The mixture then enters the regenerator where CO₂ is removed from the particles and the clean sorbent particles are circulated back to the riser.

Fig. 2 shows the entire geometry of the riser section simulated using multiphase CFD models, and Fig. 3 shows the lower section of the riser. As shown in Fig. 3, the flue gas enters from the bottom and flows upward. The solid sorbent particulates enter through the angled section, flow downward and are entrained in the flue gas and rise upward mixing with the flue gas and adsorbing CO₂. The multiphase flow, solid sorbent particles and flue gas, rise upward through the riser section and enter at the bottom of the ICFB, as shown in Figs. 4 and 5. Fig. 4 shows the entire ICFB geometry, Fig. 5 shows the lower portion, and Fig. 6 shows the mid-section. In Fig. 5, the top of the riser pipe is shown entering the lower section of the ICFB with the top of the riser ending at the bell-shaped section shown in Fig. 6. The particulates and flue gas rise upward through the ICFB and impinge upon the semi-circular diverter shown more clearly in Fig. 6. The particulates and flue gas mix, with the CO₂ being further adsorbed from the flue gas.
gas into the sorbent particles, and the particulates flow downward within the ICFB outer section. The CO₂ laden particulates settle in the bottom of the ICFB and exit through the angled piping shown on the lower left of Fig. 5. The particulates then flow into the regenerator section. The regenerator uses process steam to heat the sorbent back to a temperature in which they liberate the CO₂ adsorbed in the riser and ICFB. The CO₂ steam is separated from the flue gas; the CO₂ is sent to a sequestration system and the flue gas flows up the stack. The solid particulates settle to the bottom of the regenerator and are then sent back into the riser section and sent through the system again to capture more CO₂.

Various meso-porous materials are being considered for the sorbent particles of the carbon capture system. In the simulations presented in this paper a silica-supported amine sorbent material developed at NETL is modeled [5,6]. The CFD models use a one dimensional lumped kinetic model developed at NETL, which is a function of the local partial pressure of CO₂ and water and the surface concentration of the reactants and products of the reactions of CO₂ with amine.

3. Theory

The FLUENT® and BARRACUDA CFD codes are used to model the multi-physics of the gas and particle phases of the solid sorbent reactor. FLUENT® has both Eulerian–Eulerian (EE) and Eulerian–Lagrangian multi-phase modules; while BARRACUDA uses only an Eulerian–Lagrangian modeling method. In both codes an Eulerian description is used to model the gas phase. With the Eulerian description the gas phase is approximated as a continuum and the temperature, and density fields are solved using the appropriate conservation and constitutive laws. In FLUENT®’s Eulerian–Eulerian multi-phase model the particle phase is also modeled as a continuum with its own velocity, temperature and density fields. While in the Eulerian–Lagrangian models of FLUENT® and BARRACUDA a discrete Lagrangian phase is used to model the particle phase.

3.1. Eulerian–Eulerian

One method of flow solution involves using an Eulerian–Eulerian multiphase computational model to predict the flow of the continuous gaseous and dispersed solids phases. The principal difference of the multi-phase model compared with a single phase model is the appearance of the volume fraction for each phase, as well as terms that model the exchange of momentum and mass transfer between the phases [7]. The equations detailed below are applicable to the Eulerian–Eulerian multi-phase model in which the secondary granular phase is treated as continua [8]. In the current computations, the primary carrier phase is defined by the gaseous phase, while the secondary granular phase is defined by the micron sized solid sorbent particles.

The mass balance for each phase is given by:

$$\frac{\partial}{\partial t} \left( \alpha_q \rho_q \right) + \nabla \cdot \left( \alpha_q \rho_q \vec{v}_q \right) = 0$$

(1)

where $\rho_q$ is the density, $\alpha_q$ is the volume fraction and $\vec{v}_q$ represents the velocity of phase $q$. Within the control volume, the volume fraction
occupied by each phase cannot be occupied by other phases and the summation of volume fractions must equal unity. Momentum transport for each phase is given as [9]:

\[
\frac{\partial}{\partial t} \left( \alpha_s \rho_s \vec{v}_s \right) + \alpha_s \rho_s \vec{v}_s \cdot \nabla \vec{v}_s = -\alpha_s \nabla p + \alpha_s \rho_s \ddot{g} + \nabla \cdot \mathbf{T}_s + \sum_{q=1}^{n} K_{pq} (\vec{v}_q - \vec{v}_p) + \vec{F}_s
\]

where \( p \) is the pressure, \( \mathbf{T}_s \) is the stress tensor of the qth phase, \( \ddot{g} \) is the gravity force, and \( \vec{F}_s \) represents the external force. \( K_{pq} \) represents the interaction between the fluid and the solid phases.

### 3.1.1. Fluid phase stress tensor

The stress tensor for the fluid phase (subscript \( f \)), resulting from the forces experienced by the fluid as a result of contact with particles is given as [9]:

\[
\mathbf{T}_f = \alpha_f \mu_f \left( \nabla \vec{v}_f + \nabla \vec{v}_f^T \right) + \alpha_f \left( \lambda_f - \frac{2}{3} \mu_f \right) \nabla \cdot \vec{v}_f \mathbf{I}
\]

where \( \alpha_f \) is the volume fraction of fluid phase, \( \lambda_f \) and \( \mu_f \) are the bulk and shear viscosities and \( \mathbf{I} \) is the unit tensor.

### 3.1.2. Solids phase stress tensor

Forces experienced by the particles as a result of contact with other particles give rise to the stress tensor for the granular phase (subscript \( p \)), which can be written mathematically as [9]:

\[
\mathbf{T}_p = -p_s \delta + \alpha_p \mu_p \left( \nabla \vec{v}_p + \nabla \vec{v}_p^T \right) + \alpha_p \left( \lambda_p - \frac{2}{3} \mu_p \right) \nabla \cdot \vec{v}_p \mathbf{I}
\]

where terms in the above equation with subscript \( s \) are corresponding terms for the solids phase, \( p_s \) is the solids pressure and \( \delta \) is the Kronecker delta. The solids stress tensor contains shear and bulk viscosities arising from particle momentum exchange due to translation and collision and are represented as \( \mu_p \) and \( \lambda_p \) in the above equation. While the solids shear viscosity is formed by adding the collisional and kinetic components, the solids bulk viscosity accounts for the resistance of the solids to compression and expansion and are modeled based on the formulations proposed by [7]. The solids pressure \( p_s \) represents the inter-particle pressure which is due to the collision of the particles amongst themselves or with the surrounding walls. The solids pressure is calculated independently and used for the pressure gradient term (\( \nabla p_s \)) in the secondary phase momentum equation. The solids pressure \( p_s \) consists of a kinetic term and a second term due to particle collisions and is given as

\[
p_p = \alpha_p \rho_p \theta_s + 2 \rho_p (1 + e_w) \alpha_p^2 g_{0.35} \theta_p
\]

where, \( \theta_s \) is the granular temperature which is derived from the kinetic theory of gasses and is proportional to the kinetic energy of the fluctuating particles, \( e_w \) is the coefficient of restitution which accounts for the inelastic or elastic collisions and \( g_{0.35} \) represents the radial distribution function,

\[
g_{0.35} = \left[ 1 - \left( \frac{\alpha_p}{\alpha_p \max} \right) \right]^{-1}
\]

which is a correction factor that modifies the probability of collisions between the solid particles when the granular phase becomes dense.

The granular temperature model is based on the kinetic theory model and is proportional to the kinetic energy of the solid (particulate) phase [19]

\[
\frac{3}{2} \left[ \frac{\partial}{\partial t} \left( \rho_s \alpha_s \Theta_s \right) + \nabla \cdot \left( \rho_s \alpha_s \vec{v}_s \Theta_s \right) \right] = \left( -p_s \mathbf{I} + \mathbf{F}_s \right) : \nabla \vec{v}_s + \nabla \cdot \left( k_3 \nabla \Theta_s \right) - \gamma_0 \alpha_s \Theta_s \left( \Theta_s - \Theta \right)
\]

where

\[
\left( -p_s \mathbf{I} + \mathbf{F}_s \right) : \nabla \vec{v}_s \quad \text{the generation of energy by the solid stress tensor} \]

\[
k_3 \nabla \Theta_s \quad \text{the diffusion of energy} \quad \left( k_3 \text{ is the diffusion coefficient} \right)
\]

\[
\gamma_0 \alpha_s \Theta_s \left( \Theta_s - \Theta \right) \quad \text{the collisional dissipation of energy}
\]

\[
\phi_{\text{ex}} \quad \text{the energy exchange between the} \quad \text{Ith fluid phase and the} \quad \text{sth solid phase.}
\]

We used the Gidaspow et al.’s model of the diffusive flux of granular energy, \( k_3 \nabla \Theta_s \), where \( k_3 \) is given by [9]:

\[
k_3 = \frac{150 \rho_s d_s \sqrt{(\rho g)_s}}{384 (1 + e_w) (g_{0.35})} \left[ 1 + 6 \left( \rho_s g_{0.35} (1 + 2 e_w) \right)^2 \right]^{1/2} + 2 \mu_s \alpha_s^2 d_s (1 + e_w) g_{0.35} \sqrt{\frac{\Theta_s}{\pi}}
\]

and the Lun et al.’s expression for the collisional dissipation of energy, \( \gamma_0 \), [9]:

\[
\gamma_0 = \frac{12}{d_s \sqrt{\pi}} \rho_s \alpha_s^2 \Theta_s^{3/2}
\]

The energy exchange between the fluid and the solid phases is given by

\[
\phi_{\text{ex}} = -3 k_3 \Theta_s
\]

We used the algebraic formulation option in FLUENT®, which neglects the advection and diffusion in the above equation.

### 3.1.3. External forces

Solid particles entrained in the flow are subject to drag by the surrounding fluid. This results in a momentum exchange, which is given as [7],

\[
K_{nf} = \frac{3}{4} C_D \frac{\alpha_p \rho_p \mu_p}{d_p} \left( \frac{\vec{v}_p - \vec{v}_f}{\Delta t} \right)^{2.65}
\]

where \( d_p \) represents the diameter of the particles in the secondary phase, which in the current simulations are comprised of mono-dispersed particles and \( C_D \) represents the drag coefficient on a single sphere which in turn is related to the relative particle Reynolds number by

\[
C_D = \frac{24}{\alpha_p \nu_{Re_p}} \left[ 1 + 0.15 \left( \alpha_p \nu_{Re_p} \right) \right]^{0.87}
\]

where

\[
\nu_{Re_p} = \frac{\rho_p d_p \left( \vec{v}_p - \vec{v}_f \right)}{\mu_f}
\]

The effects of turbulence are modeled using a per-phase \( k-e \) Reynolds Averaged Navier Stokes (RANS) model. Coupling between primary carrier and secondary particle phases is achieved via the inter-phase momentum transfer term. All the sorbent particles are approximated as smooth spheres in the current computations. The current work does
not account for any particle size changes due to absorption and desorption of CO$_2$.

3.2. Eulerian–Lagrangian

In the Eulerian–Lagrangian models, a Lagrangian description is used for the particle (dispersed) phase with a Eulerian description of the gas phase. In FLUENT® the dense discrete phase method (DDPM) is used to model the solid phase of the Eulerian–Lagrangian model, which, in contrast to a discrete phase model (DPM) [10], accounts for the volume excluded by the particles in the gas phase conservation equations. In BARRACUDA, the multi-phase particle-in-cell (MP-PIC) method [11] is used to model the particle phase.

3.2.1. FLUENT® dense discrete phase model

The formulation for the FLUENT® DDPM module uses a modified Eulerian description of the gas phase which accounts for the volume fraction ($\alpha$) of the DDPM particle phase. The modified mass conservation for the gas phase is,

$$\frac{\partial}{\partial t} (\rho_f \alpha_f) + \nabla \cdot (\rho_f \alpha_f \mathbf{v}_f) = (m_{mf} - m_{mp}) + S_f$$

(14)

and for the particle phase is,

$$\frac{\partial}{\partial t} (\rho_p \alpha_p) + \nabla \cdot (\rho_p \alpha_p \mathbf{v}_p) = (m_{fp} - m_{mp}) + S_p.$$  

(15)

The momentum conservation for the gas phase is,

$$\frac{\partial}{\partial t} (\rho_f \mathbf{v}_f) + \nabla \cdot (\rho_f \mathbf{v}_f \mathbf{v}_f) = -\alpha_f \nabla p_f + \nabla \cdot [\alpha_f \mu_f (\nabla \mathbf{v}_f - \nabla \mathbf{v}_f)] + \alpha_f \mathbf{g} + F_f + (K_{pf} (\mathbf{v}_f - \mathbf{v}_p) + m_{fp} \mathbf{v}_{pf} - m_{mp} \mathbf{v}_{mp}) + K_{DPM} (\mathbf{v}_{DPM} - \mathbf{v}_p) + S_{DPM explicit}$$

(16)

and for the particle phase is,

$$\frac{\partial}{\partial t} (\rho_p \mathbf{v}_p) + \nabla \cdot (\rho_p \mathbf{v}_p \mathbf{v}_p) = -\alpha_p \nabla p_p + \nabla \cdot [\alpha_p \mu_p (\nabla \mathbf{v}_p - \nabla \mathbf{v}_p)] + \alpha_p \mathbf{g} + F_p + (K_{fp} (\mathbf{v}_p - \mathbf{v}_f) + m_{fp} \mathbf{v}_{pf} - m_{mp} \mathbf{v}_{mp})$$

(17)

where $p$ denotes the particle phase and $f$ denotes the gas phase, $\alpha_0$ is the volume fraction of phase $p$, $\rho_p$ is the density of phase $p$, $\mathbf{v}_p$ is the velocity of $p$, $p_p$ is the pressure of $p$, $m_{pf}$ is the mass transfer from phase $p$ to phase $f$, $K_{pf}$ is the interphase momentum exchange coefficient, $S_p$ is an optional mass source term, $\mu_p$ is the shear viscosity, and $F_p$ is an external force. $\mathbf{v}_{pf}$ is the interphase velocity which is defined as either $\mathbf{v}_p$ or $\mathbf{v}_f$ depending on $m_{pf}$. $K_{fp}$ is calculated from the volume fraction of the phases and the drag force ($f$) as,

$$K_{pf} = \frac{\alpha_f \mu_f d_p^2}{18 \tau_s}$$

(18)

where $\tau_s$ is,

$$\tau_s = \frac{\rho_p d_p^2}{18 \mu_p}$$

(19)

where $d_p$ is the diameter of the particles.

The pressure in the solid particles is calculated from,

$$p_p = \alpha_p \rho_p \Theta_p + 2 \rho_p (1 + e_{pp}) \alpha_p^2 \sigma_{app} \Theta_p$$

(20)

where $e_{pp}$ is the coefficient of restitution for particle collisions, $\sigma_{app}$ is the radial distribution function and $\Theta_p$ is the granular temperature. $\sigma_{app}$ governs the transition from a compressible condition where particles can be packed closer together to an incompressible condition where $\alpha = \alpha_{max}$; $\sigma_{app}$ can also be interpreted as the non-dimensional distance between particles,

$$\sigma_{app} = \left[1 - \left(\frac{\alpha}{\alpha_{max}}\right)^2\right]^{-1}.$$  

(21)

The solids shear viscosity of the particles is calculated from the collisional, kinetic, and (optional) frictional components,

$$\mu_p = \mu_p\text{col} + \mu_p\text{kin} + \mu_p\text{fr}$$

(22)

where,

$$\mu_p\text{col} = \frac{4}{5} \alpha_p \rho_p d_p \sigma_{pp} (1 + e_{pp}) \left(\frac{\Theta_p}{\bar{\rho}_p}\right)^{1/5} \alpha_p$$

(23)

$$\mu_p\text{kin} = \frac{\alpha_p d_p \rho_p \sqrt{\Theta_p}}{6(1 - e_{pp})} \left[1 + \frac{2}{3}(1 + e_{pp}) (3e_{pp} - 1) \alpha_p \sigma_{pp}\right]$$

(24)

and $\mu_p\text{fr}$ is not included unless the particles are densely packed and thus not enabled in these simulations. The bulk viscosity of the particles is calculated as,

$$\lambda_p = \frac{2}{3} \alpha_p \rho_p d_p \sigma_{pp} (1 + e_{pp}) \left(\frac{\Theta_p}{\bar{\rho}_p}\right)^{1/5}.$$  

(25)

The granular temperature in the particles, $\Theta_p$, is calculated based on velocity, volume fraction, etc.

The trajectory of the particles is calculated by a force balance

$$\frac{d\mathbf{v}_p}{dt} = F_D (\mathbf{v}_p - \mathbf{v}_f) + \frac{g_s (\rho_p - \rho_f)}{\rho_p} + \mathbf{F}_{\text{interaction}}$$

(26)

where $F_D (\mathbf{v}_p - \mathbf{v}_f)$ is the drag force per unit particle mass and is,

$$F_D = \frac{18 \rho c_\text{d} \text{Re}}{24 \rho d_p \overline{\mu}_p}$$

(27)

where the Reynolds number is,

$$\text{Re} = \frac{\rho_p d_p |\mathbf{v}_p - \mathbf{v}_f|}{\mu_p}$$

(28)

$\mathbf{F}_{\text{interaction}}$ models the additional acceleration on a particle due to interparticle interactions and is defined as,

$$\mathbf{F}_{\text{interaction}} = -\frac{1}{\rho_p} \mathbf{v}_p \cdot \overline{\mathbf{F}}$$

(29)

The conservation equation for the granular temperature, $\Theta_p$, is solved using the average particle velocity field, and is analogous to the granular temperature/kinetic theory for the Eulerian–Eularian solid phase $\Theta_0$ described in Section 3.1.2.

3.2.2. BARRACUDA

CFD’s BARRACUDA code solves the fluid and particle equations in three dimensions using the averaged Navier–Stokes equations. Strong coupling of the fluid phase with the particle phase is obtained by reviewing work by [12,13] as well as the original references cited in these two papers. The particle momentum equation uses the multi-phase particle-in-cell (MP-PIC) formulation of Andrews and O’Rourke [14] and Snider [11] including a relaxation-to-the-mean term to represent damping of particle velocity fluctuations due to particle
collisions [15]. Mass, momentum, and energy of the two-phase mixture are conserved by exchange terms in the gas phase mass, momentum, and energy equations, respectively.

The mass and momentum equations for the fluid-phase are averaged forms of the detailed fluid-phase mass and momentum equations of Anderson and Jackson [16] and Jackson [17],

$$\frac{\partial (\alpha_i \rho_i)}{\partial t} + \nabla \cdot (\alpha_i \rho_i \mathbf{u}_i) = \delta \dot{m}_p$$

(30)

and

$$\frac{\partial (\alpha_i \rho_i \mathbf{u}_i)}{\partial t} + \nabla \cdot (\alpha_i \rho_i \mathbf{u}_i \mathbf{u}_i) = -\nabla P + F + \alpha_i \mathbf{D} + \nabla (\alpha_i \tau_f)$$

(31)

where \(\alpha_i\) is the fluid volume fraction, \(\rho_i\) is fluid density, \(\mathbf{u}_i\) is the fluid velocity vector, \(\delta \dot{m}_p\) is the gas mass production rate per volume from gas-particle chemistry, \(P\) is the mean flow gas thermodynamic pressure, \(F\) is the inter-phase momentum transfer rate per unit volume, \(g\) is the gravitational acceleration and \(\tau_f\) is the fluid stress tensor.

The constitutive equation for the non-hydrostatic part of the stress, \(\tau_f\), is

$$\tau_{f i j} = \mu \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) - \frac{2}{3} \mu \delta_{i j} \frac{\partial u_i}{\partial x_j}$$

(32)

where \(i\) and \(j\) are directions and \(\mu\) is a shear viscosity, which is the sum of the laminar shear viscosity and a turbulence viscosity. Large eddies are calculated with the unresolved sub-grid turbulence being modeled with a turbulent or commonly referred to as an eddy-viscosity which is given by

$$\mu_t = C \rho_f \Delta^2 \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right)^2$$

(33)

where \(C\) is the Smagorinsky’s [18] coefficient (0.01) and \(\Delta\) is the sub-grid length scale which is the cubic root of the sum of the product of orthogonal distances across a calculation cell.

Transport equations are solved for each gas species with the total fluid phase properties calculated from the mass fractions \(Y_{f i}\) of the gas species \(i\). Mass transferred between gas species through the breaking and forming of chemical bonds is represented as chemical source terms \(\delta \dot{m}_i\) in the individual gas species transport equations,

$$\frac{\partial (\alpha_i \rho_i Y_{f i})}{\partial t} + \nabla \cdot (\alpha_i \rho_i \mathbf{u}_i Y_{f i}) = \nabla \cdot (\rho_i D \alpha_i \nabla Y_{f i}) + \delta \dot{m}_i$$

(34)

where \(D\) is the turbulent mass diffusivity and is related to the viscosity by the Schmidt number correlation,

$$\frac{\mu}{\rho_f D} = Sc.$$  

(35)

Eqs. (30) and (31) are the mass and momentum balances for the gas mixture, respectively, where the gas mixture properties are based on the mass fractions of the gas species, solved for using Eq. (34). Furthermore, the flow is compressible, and the gas phase pressure, density, and mass fractions are related through an equation of state which is the ideal gas equation in CPFD’s BARRACUDA. Using the ideal gas equation of state, the partial pressure of gas species \(i\) is

$$P_i = \frac{\rho_i Y_{f i} R T}{M_{W_i}}$$

(36)

where \(R\) is the universal gas constant, \(T_f\) is the gas mixture temperature, and \(M_{W_i}\) is the molecular weight of gas species \(i\) and the total mean flow gas thermodynamic pressure is

$$P = \sum_{i=1}^{N_f} P_i$$

(37)

In CPFD’s BARRACUDA as well as in other MP-PIC method codes, the dynamics of the particle phase is predicted by solving a transport equation for the particle distribution function (PDF), \(f\). This function is based upon the work of [14], BARRACUDA assumes that \(f\) is a function of particle spatial location \(x_p\), particle velocity \(u_p\), particle mass \(m_p\), particle temperature \(T_p\), and time \(t\). Thus,

$$f = f(x_p, u_p, m_p, T_p, t)$$

(38)

and \(N_p\) is the average number of particles per unit volume with velocities in the interval \((u_p, u_p + du_p)\), masses in the interval \((m_p, m_p + dm_p)\), and temperature in the interval \((T_p, T_p + dT_p)\) and is given by

$$N_p = f(x_p, u_p, m_p, T_p, t) dm_p dm_p dT_p.$$  

(39)

In CPFD’s BARRACUDA, MP-PIC code, the fluid mass source term from Eq. (30) is

$$dm_p = -\iiint f \frac{dm_p}{dt} dm_p dm_p dT_p$$

(40)

where the time-rate-of-change of particle mass \(dm_p/dt\) is the rate of change of the particle mass producing gasses through chemistry.

In BARRACUDA, the acceleration on a particle is

$$\frac{dm_p}{dt} = D_p (u_j - u_p) - \frac{1}{\rho_p} \nabla P - \frac{1}{\alpha_p \rho_p} \nabla \tau_p + g + \frac{\tau_f - u_p}{\Delta}$$

(41)

where \(\alpha_p\) is the solids volume fraction; \(\rho_p\) is the solid material mass density; \(D_p\) is the drag function which depends on particle size, velocity, position, and time; \(\tau_p\) is the solids contact stress, which depends on spatial location; \(u_p\) is the local mass-averaged particle velocity; and \(\tau_{f \alpha}\) is a particle collision damping time from [15].

The solids volume fraction is related to the PDF \(f\) by

$$\alpha_p = \iiint f \frac{dm_p}{P_p} dm_p dm_p dT_p.$$  

(42)

and the fluid volume fraction is

$$\alpha_f = 1 - \alpha_p.$$  

(43)

The particle force per volume on the fluid phase in Eq. (31) is

$$F = -\iiint \left[ \frac{m_p}{P_p} D_p \left( u_j - u_p \right) - \nabla P \right] dm_p dm_p dT_p.$$  

(44)

The equation for solid displacement is

$$\frac{dx_p}{dt} = u_p.$$  

(45)

3.3. Chemical reactions

The CO₂ absorption process is modeled using a homogenous reaction model [5]. The original model was written in terms of site-fractions and has been converted to a mass basis for inclusion in the various CFD codes. Only the dry absorption step is included in this model.
The dry absorption process is modeled as the reaction of gaseous CO₂ with two amine (R₂NH) sites to produce a carbamate ion (R₂NCO₂⁻) and pronated amine ion (R₂NH₂⁺):

\[ 2R₂NH + CO₂ \rightarrow \frac{k_{f}}{k_{i}} R₂NCO₂⁻ + R₂NH₂⁺. \]  

(46)

The reaction rate (mol-CO₂ absorbed/m³/sorbent/s) takes an Arrhenius form:

\[ RR = -k \left( \frac{[CO₂][R₂NH²]}{[R₂NCO₂⁻][R₂NH₂⁺]} \right) \text{kmol/s/m³ solid} \]  

(47)

where the forward rate and equilibrium constants take the following form:

\[ k = AT_s R_g \exp \left( \frac{-ΔH_f}{T_s} \right) \]  

(48)

\[ K = \frac{R_s T_s}{P_{ref}} \exp \left( \frac{dΔH_f}{T_s} \right). \]  

(49)

The simulations were performed at constant temperature so the difference between the solid temperature and gas temperature was not taken into effect.

The CO₂ concentration, [CO₂] is per unit volume-gas and the solid species concentrations are per-unit volume solid. To get the reaction rate in an arbitrary volume (Eulerian form) Eq. (47) needs to be multiplied by the solid volume fraction, leading to

\[ RR_{EE} = -ak \left( \frac{[CO₂][R₂NH²]}{[R₂NCO₂⁻][R₂NH₂⁺]} \right) \text{kmol/s/m³ space}. \]  

(50)

For the Lagrangian models Eq. (47) is multiplied by the particle volume (V_p) to get the reaction rate on a per-particle basis

\[ RR = V_p \left( \frac{[CO₂][R₂NH²]}{[R₂NCO₂⁻][R₂NH₂⁺]} \right) \text{kmol/s/particle}. \]  

(51)

4. Computational meshing and geometry simplifications

Due to the complexity and scale of the 1 kWe solid sorbent carbon capture system, simplifying assumptions and modifications were made to the geometry in both the FLUENT® and BARRACUDA simulations. In the following sub-sections we briefly discuss these changes.

4.1. FLUENT® meshes

With the available computational resources at our disposal and given the complexity of the 1 kWe reactor, it was decided to break up the reactor into smaller individual components, which could be easily managed for computations, for the FLUENT® EE and DDPM simulations. With this in mind, components that play a significant role in the carbon capture process were identified for the purpose of computations and were split into the lower riser section (modeled as 3D), the upper riser section (modeled as axi-symmetric 2D), and the ICFB (modeled as 3D). Simulations for obtaining an Eulerian–Eulerian multiphase flow solution for each of these sections were performed using FLUENT® and the flow and numerical parameters were kept uniform across the simulations to maintain consistency in the results. Coupling between each section was accomplished by using the average output from one section as the input to the next section, i.e. the output of the bottom riser section was used as the input for the top riser section. For the FLUENT® DDPM simulations only the 3D bottom riser section was considered due to computational issues encountered with DDPM as will be discussed in Section 6.3.

A grid was created for each of the sections described previously, i.e. the 3D riser bottom, the 2D axi-symmetric riser and the 3D ICFB, using Gambit®. To preserve the geometry from being distorted during grid generation, a combination of structured and unstructured grids was used in creating the 3D geometry, while the 2D riser was meshed using a structured triangular mesh. Various meshes were created to measure grid size dependence of the mesh and to compute grid effects and computing time required for arriving at the solution. The original meshes considered for the EE simulations included a 3D mesh of the entire riser section (3D bottom section plus the upper axi-symmetric section). Based on preliminary studies, the riser was divided into the 3D and 2D sections; with a mesh size of 51490 cells for the lower 3D riser section as shown in Fig. 7, and a mesh size of 2000 cells for the 2D axi-symmetric section. The 2D structured mesh was created using 5 grid points along the radial direction and 400 grid points along the central vertical axis. The length of the 3D bottom section of the riser is approximately 1 m, while the length of the axi-symmetric 2D section is approximately 9 m.

For the ICFB, which is approximately 5 m-long, a Gambit® mesh consisting of ~450,000 cells is used as shown in Fig. 8. Due to the complex geometry of the ICFB it was not possible to simplify or divide it into smaller sections, which lead to long computational times in the ICFB.
For the DDPM simulations a cut cell mesh was used to model the entire riser section. Initially the Gambit® mesh of the entire riser section, which was used in the initial EE simulations, was used for the DDPM simulations; however, convergence issues and instability lead to the use of a cut cell mesh for the DDPM simulations. The DDPM simulations can become unstable when using a computational mesh with a large ratio of maximum to minimum cell volume. In the original Gambit® mesh of the entire riser section this ratio was 64 and the mesh contained over 226,000 cells. To overcome the stability issues a cut cell mesh with a ratio of 30 was used for the DDPM simulations. The cut cell mesh also reduced the mesh size for the riser to 31,092 cells.

4.2. BARRACUDA mesh

BARRACUDA is a serial code using a cut cell meshing technique to model complex geometries with a Cartesian grid. For the 1 kWe carbon capture system the whole reactor loop (riser and ICFB) was modeled as a single unit. The geometry of the 1 kWe system was simplified to allow for the cut cell mesh. In the riser the angled pipe where the solid particles enter the system was not modeled; instead an inlet boundary condition was applied where the angled pipe would have been. In the ICFB the geometry was cut off above the diffuser plate to reduce the mesh size. The diffuser plates stop particles from entering the upper region of the ICFB and there is no significant flow of flue gas in the upper region.

In BARRACUDA’s cut cell meshing technique simplifications to the mesh are made based on the aspect ratios of the cells and user defined criteria. Small cells with large aspect ratios are either dropped or added to neighboring cells depending upon the gridding user defined criteria; these cells are then considered null cells. Therefore, for the model of the entire 1 kWe system (riser to the bottom of the diffuser plate in the ICFB) the total of real and null cells is 2,587,200. For the simulation run, there are 696,638 real cells; in the null cells no calculations are preformed; however they do consume memory. Fig. 9 shows the overall cut cell mesh for the system. A more detailed view of the grid for the bottom of the riser, including the expansion and contraction, is shown in Fig. 10. The grid for the top of the riser and ICFB is detailed in Fig. 11.

Some simplifying assumptions and compromises were made to generate the grid with only 696,638 real cells. These can be seen to some extent and are discussed with respect to Fig. 5. In the ICFB, the riser top extends into the bottom of the ICFB and up into the draft tube as can be seen. In the BARRACUDA simulations the wall thickness of both the riser and draft tube has been increased so that they can get captured in the cut cell grid without increasing the cell count. In other words, a much finer grid would have to be used to capture these internal structures if the wall thickness criteria were not relaxed. Gridding and the resulting cell size have a direct impact on the calculations, model assumptions and the simulation run time. Due to the nature of the Eulerian–Lagrangian simulation method used in BARRACUDA, several of the particle clouds of the solid sorbent particles need to be able to fit within the smallest cell in the mesh. If this is not so, cells will over pack and the simulation will fail. In order to overcome this issue, either the grid size needs to be increased or the number of particle clouds needs to be increased (which will decrease the cloud size). Increasing the number of particle clouds increases the time for each calculation as more particles are tracked; in the BARRACUDA simulations presented in this paper the grid size was increased to overcome this problem, which leads to the increased wall thicknesses in the ICFB.
5. Operating conditions

The sorbent used in the 1 kWe reactor and also in the numerical simulations was developed at NETL and is referred to as 196C. It is an amine based sorbent which is impregnated on a silica substrate with a ratio of 44% sorbent to 56% silica. The bulk density and mean diameter of the solid sorbent particles are 1120 kg/m³ and 180 μm respectively. A solids circulation rate of 31 lb/h, which equates to approximately 5 g/s is used in the simulations. The flue gas circulation rate of 40 lb/h, which in turn equates to around 2.3 g/s is used for the primary carrier phase. Solids are injected from the angled inlet into the riser, while flue gas is injected from the bottom of the riser using the mass flow rates mentioned above. The composition of flue gas used in the current simulations stands at 75% N₂, 15% CO₂ and 10% O₂ by mass.

6. Comparison of model results in the riser section

Both FLUENT® and BARRACUDA were used to model the riser section of the 1 kWe solid sorbent system. The following sub-sections discuss the details of the simulations in the riser. Note that the BARRACUDA simulation models the entire loop in one simulation; for ease of comparison we break the loop into the rise and ICFB sections for post processing and for comparisons with the FLUENT® simulations.
6.1. FLUENT® Eulerian–Eulerian

FLUENT® EE simulations were carried out for the 3D lower riser section and the upper 2D riser using an initial solids volume fraction of 0.35. Initial simulations were carried out using a time step of 1e-4 s and during startup the gas velocity at the inlet was gradually ramped up from 10% to 100% of the operating velocity to achieve fast convergence. Once the simulation displayed convergence, the time step was gradually increased to 1e-3 s to reduce the computational time required for arriving at the results. Simulations were initially carried out with the k-e turbulence model, but as the simulations progressed, the turbulence model was found to introduce instability in the convergence of results. Consequently, the latter FLUENT® EE (and DDPM) simulations were performed using the Laminar flow model. To investigate the effect of varying simulation parameters on the results, EE simulations were carried out using two different built-in FLUENT® drag models—Gidaspow and Energy Minimization Multi-Scale (EMMS). The EMMS drag model is not built-in to the FLUENT® multi-phase models and was implemented via a user defined function based on [19]. Following initial studies of the drag models, it was decided to restrict all EE FLUENT® simulations to the Gidaspow drag law.

The results of the FLUENT® EE simulations are restricted to the distribution of particles in the lower 3D section and CO2 absorption in the upper 2D axi-symmetric section of the riser. Time distributions of solids within the lower 3D riser at \( t = 65, 69 \) and \( 72 \) s are shown in Fig. 12(a), (b) and (c) respectively. The solids are found to accumulate within the lower section of the riser and dissipate as they move along the length of the riser. The contour plots of Fig. 12 show that the volume fraction of particles remains relatively steady from \( t = 65 \) s to \( t = 72 \) s with slight variations in the particle distributions.

The velocity vector plots of Fig. 13 indicate a mixing zone at the angled inlet section where the solids enter the riser. As seen in the close up of the angled pipe joining the riser, the solids velocity magnitude is found to increase in the mixing zone and then reduce as it moves along the length of the riser. The velocity of particles within the full riser is observed to be in the range of approximately 2.5–3.5 m/s, with an exception near the mixing zone, where the solids are observed to accelerate to higher speeds of approximately 10 m/s.

In addition to the hydrodynamics, the reactions of the solid sorbent particles with the flue gas were also simulated in the FLUENT® EE
model. Based on the dry sorbent chemistry developed at NETL and discussed in Section 3.3, an UDF was written in FLUENT® and compiled into the existing multiphase EE model via UDF routines. The chemistry studies using the FLUENT® EE method are restricted to the upper 2D axi-symmetric riser section and the ICFB simulations. This simplification was made to reduce the computational time for the simulations. As shown in the BARRACUDA simulations the reactions occur in the lower regions of the riser which is supported by experimental data. Implementing the reactions in the FLUENT® EE model caused stability and run time issues in the lower riser section, and so the simpler 2D riser section was used to investigate the reaction implementation in FLUENT®. The 2D FLUENT® simulations were used as an initial case to overcome stability issues. To obtain a more accurate solution a 2D slice through the riser is more appropriate. Fig. 14 shows the mass fraction of CO₂ remaining in the flue gas following absorption along the length of the riser from 470 to 500 s. The values are measured at three different heights (3.8 m, 6.7 m and 9.6 m) along the length of the riser. The mass fraction distribution of the solids within the 2D section of the riser was analyzed and it revealed a predominantly dilute flow of solids, with solids volume fraction remaining below 5% in the majority of the riser, except near the bottom inlet. A snapshot of the contours of volume fraction of solids obtained by the EE simulations at a region approximately 0.63 m above the bottom end of the 2D riser at t = 500 s is shown in Fig. 15.

6.2. BARRACUDA Eulerian–Lagrangian

The results from the BARRACUDA simulation of the riser are shown in Figs. 16 through 20. The time step for the BARRACUDA simulations was generally on the order of ~2×10⁻⁴ s, varying slightly about this value. This gave a calculation time about 1 s of simulated time per week of run time. Note that this is a serial code and run on CPFD’s beta version of the code. They have since made some improvements which are in the latest commercial release. The simulation was run under isothermal conditions at 300 K. The low operating temperature eliminated any desorption of the CO₂ from the solid sorbent providing nearly complete removal of the CO₂. Future studies are incorporating the thermal effects as well as investigating the effect of both the solids and gas flow rates.

Figs. 16–18 show the conditions at the bottom of the riser. It should be noted that it took approximately 7 s to reach a pseudo-steady condition. These results are presented for a time at about twice that or 14 s.

Fig. 15. Volume fraction of solids within the riser at t = 500 s.

Fig. 16. Tracer mass fraction of adsorbed CO₂.

Fig. 17. Tracer mass fraction of adsorbent.
The solid consists of the substrate (silica), the adsorbent (amine) and the carbonated material (adsorbed CO$_2$). The material fed to the riser at the lower left of the expanded area is 50% substrate, 45% adsorbent and 5% adsorbed CO$_2$ to reflect that incomplete regeneration might occur. In Fig. 16, it can be seen that the 5% adsorbed CO$_2$ sorbent enters at the lower left at a mass fraction of 45%. As the particles travel up through the riser, they react, causing the decrease in the unreacted sorbent mass fraction. As a note, it can be seen that some particles pass through the riser without adsorbing any CO$_2$. This might indicate that the solids flow rate is too high for the CO$_2$ feed rate.

Fig. 18 presents the particulate velocities in the riser. Solids are moving down on the right side and up at the left side. Typical solids velocities are around 2 m/s up and 2 m/s down.

Simulation results for the CO$_2$ mass fraction in the gas at $t = 12$, 13 and 14.28 s are shown in Figs. 19 and 20. The results presented in Fig. 19 for the simulation show that the CO$_2$ mass fraction is essentially zero by the time the gas exits the lower riser. This is quantified as the average value of the CO$_2$ mass fraction and is plotted in Fig. 20 as a function of the height.

The solids are carried up the riser. Note the change in particle color from dark blue to light blue and from green to yellow such that the mass fraction of some particles will be near 75% adsorbed CO$_2$. The behavior in Fig. 17 for the unreacted sorbent is nearly the opposite as that for the adsorbed CO$_2$ in Fig. 16. Particles enter at the lower left at a mass fraction of 45%. As the particles travel up through the riser, they react, causing the decrease in the unreacted sorbent mass fraction. As a note, it can be seen that some particles pass through the riser without adsorbing any CO$_2$. This might indicate that the solids flow rate is too high for the CO$_2$ feed rate.

Fig. 18. Particle velocity at bottom of the riser.

Fig. 19. Simulation results for CO$_2$ mass fraction at times of 12 s, 13 s and 14.28 s.

Fig. 20. Cross section average CO$_2$ mass fraction values at times equal to 12 s, 13 s and 14.28 s.
6.3. DDPM riser simulations

The DDPM FLUENT® model is a relatively new feature in FLUENT® and has only been available in the latest versions of FLUENT® (version 12 or later). As such, there is little documentation available on the model and its stability in modeling fluidized bed-type systems.

In the DDPM models of the 1 kWe carbon capture system, initial simulations showed instability and divergence as the solid particles began to accumulate in the system. Solving these issues has been a difficult task; issues with particle packing and subsequent divergence have been an ongoing challenge. Based on numerous discussions with FLUENT® technical support, various DDPM parameters were adjusted in an attempt to increase stability. These parameters included the following: solvers used for the governing equations, Lagrangian time step parameters, under relaxation factors, number of solution iterations, inlet boundary conditions, and turbulence model parameters.

A stable DDPM solution for the riser was finally achieved with the most significant factors affecting the solution being:

- Using a FLUENT® cut cell mesh with a lower maximum-to-minimum cell volume ratio than the Gambit® mesh. Using a cut cell mesh reduced the computational time of the DDPM simulations and reduced the ratio of maximum-to-minimum cell volume, which increases the stability of particle tracking in DDPM.
- Increasing the number of particles per DDPM cloud. In the FLUENT® DDPM model, DDPM particles are transported through the simulation...
domain as clouds of particles. By varying the inlet conditions for the particles, you can control how many particles are in each cloud. Based on FLUENT®'s suggestions, we increased the number of particles per cloud to 100, which greatly decreases the computational time for particle tracking.

- Turning off the frictional pressure model. In areas with large gradients in particle loading, the frictional pressure model can cause instability. By turning the model off, the stability is increased.
- Using the laminar viscous model. The velocity of the gas flow in the riser is low enough that the laminar viscous model should give accurate results.
- Ensuring that one inlet boundary condition is a velocity condition and another is mass flow. A numerical issue in FLUENT® requires only one mass flow boundary condition to be defined in the DDPM simulations, so one of the mass flow boundary conditions (the gas flow of the solids inlet) was converted to a velocity boundary condition.

The hydrodynamic solution of the stable DDPM model was compared with a FLUENT® EE solution with laminar flow and the same cut cell mesh as DDPM. Both the EE and DDPM simulations were initially loaded with 0.35 volume fraction of solids and used a gradually increasing mass flow rate to reach a steady state quickly.

Both the DDPM and EE simulations predict 5 g/s of solids leaving the riser at steady state (Fig. 21). However, the volume integral of the solid particle’s volume fraction in the riser predicted by the DDPM simulation is higher than that of the EE cut cell simulation (Fig. 22). A second DDPM simulation was done with 50 particles per cloud to investigate the sensitivity of the DDPM model to the number

![Fig. 23. Contour plots of the solid particles volume fraction in the EE and 100 particles per cloud DDPM simulations.](image)

![Fig. 24. Solid sorbent velocity magnitude in the ICFB.](image)
of particles per cloud. The simulation with 50 particles per cloud ran much slower than the simulation with 100 particles per cloud but predicted a volume fraction of particles closer to that of the EE simulation as seen in Fig. 22. The 50 particles per cloud case show the sensitivity of the DDPM simulation to resolution. By grouping more particles together (100 particles per cloud) you sacrifice the accuracy of the result. Using a finer resolution (50 particles per cloud) produces a more accurate result but requires much more computational time to complete the simulation.

As shown in Fig. 23, the DDPM simulation with 100 particles per cloud predicts a higher retention of solid particles in the riser than the EE cut-cell simulation. At this time it is suspected that the DDPM simulations with 100 and 50 particles per cloud are too coarse to accurately model the multi-phase flow in the riser. Due to the decreasing computational speed with an increasing number of particle clouds (decreasing particles per cloud), it is not practical or beneficial to consider finer DDPM cases.

7. Comparison of model results in the ICFB

The reactions and hydrodynamics of the ICFB were modeled using both the FLUENT® EE method and BARRACUDA’s Eulerian–Lagrangian modeling method. In the following sections the results of these simulations are discussed. The FLUENT® DDPM method was not used to
consider the ICFB. Initial simulations of the ICFB with DDPM resulted in numerous instabilities and long computational times.

7.1. Eulerian–Eulerian results

A laminar viscous model is used to simulate the hydrodynamics and chemical reactions of the gas and particles in the ICFB using the FLUENT® EE multi-phase model. To reduce the run time to steady state, the ICFB is pre-filled with 0.35 volume fraction of solid sorbent particles and the mass in-flow rates of the flue gas and the solid sorbent are gradually increased from 10% to 100% of the maximum velocity as was done in the riser FLUENT® EE simulations.

Figs. 24 and 25 show the velocity of the solid phase in the ICFB and the static pressure of the mixture. It can be seen that the velocity of the solid particles is elevated near the inlet and the diffuser plates while in the rest of the ICFB the flow appears close to zero with a gradually decreasing pressure distribution from the bottom to the top of the ICFB.

The results of the mass fraction of the amine adsorbent (non-reacted solid sorbent) and the mass fraction of the CO\textsubscript{2} in the flue gas are shown in Figs. 26 and 27. Since CO\textsubscript{2} adsorption consumes the amines, the mass fraction of CO\textsubscript{2} gradually increases as the amine adsorbent is depleted. The area-weighted average CO\textsubscript{2} mass fraction evolution at different heights is shown in Fig. 28.

**Fig. 27.** Mass fraction of CO\textsubscript{2}.

**Fig. 28.** Area-weighted average CO\textsubscript{2} mass fraction evolution at different heights.
fraction of the unreacted adsorbent decreases where carbonating reactions occur. In Fig. 26, it can be seen that 0.44 mass fraction of amines enters at the bottom inlet, and rises up in the inner pipe without any significant reduction. After being directed into the outer regions of the ICFB, the amines are gradually eliminated as the particles falling back to the bottom of the ICFB, indicating most of the CO₂ removal takes place in this region. It is also observed that there is a significant mass concentration of the unreacted sorbents in the bottom region. This might indicate that a higher flow rate is needed for the aeration ring inlets of the ICFB to enhance the fluidization. Fig. 27 shows that the CO₂ remaining in the flue gas has nearly the same distribution patterns as the amine adsorbent.

The mass fraction evolution results for CO₂ in the flue gas are shown in Fig. 28 at several different heights (11 m, 12 m, and 13 m) along the ICFB. Note the height is measured for the whole carbon adsorption system; so the bottom of the ICFB is at the height of ~9 m. The area-weighted average mass fraction of CO₂ in the flue gas is plotted against the time. Over the period shown in Fig. 28 the multi-phase flow is at steady state; however, the reactions have just been introduced to the system and so the mass fraction of CO₂ is changing due to the transient nature of the reactions. It is clear that there is less CO₂ in the gas phase at the lower locations, which indicates that most of the CO₂ adsorption happens in the bottom half of the ICFB. As the ICFB reactions approach steady state, the three curves of Fig. 28 level off.

7.2. BARRACUDA ICFB

In the BARRACUDA simulation of the ICFB, in addition to the geometrical relaxation of the wall thickness constraints, the input gas location was also relaxed such that a model could get started quickly and without adding more complexity to the gridding and the already small cells in this region. For the simulation, all gas enters the ICFB uniformly at the bottom face. This allows a greater amount of gas to bypass the central draft tube, but allows enough to get the needed recirculation in the reactor. It also allows some solids to be entrained up into the middle of the ICFB near the central draft tube. These solids quickly become de-fluidized and fall back into the bed. Figs. 29 and 30 show the volume fraction of solids and the velocity of solids in the bottom region of the ICFB. Fig. 31 shows the particle flow throughout the ICFB including the particle diffuser plate at the top of the ICFB. All three figures show a slice through the center of the ICFB as can be seen by noting the internals. Fig. 31 shows the solids velocity throughout the ICFB. The change in direction of the
solid particles as they hit the diffuser plate can be seen in Fig. 31. As the particles reach the diffuser plate their velocity becomes negative indicating their flow toward the bottom of the ICFB — note color change from light (pink) for positive velocities to dark (dark blue) for negative velocities.

8. Discussion and conclusions

The research presented in this paper has focused on simulating a 1 kWe solid sorbent carbon capture system and investigating the capabilities of state of the art CFD modeling tools to simulate the multi-phase flow in the system. We have considered various models and model settings to simulate the hydrodynamics and reactions in the system. This includes Eulerian–Eulerian and Eulerian–Lagrangian multi-phase modeling methods, various particle drag models, and different meshing methods. These simulations have provided valuable insight into the multi-physics of the carbon capture system and the strengths and limitations of the various models available in state-of-the-art commercial CFD software.

In particular, we have determined that using a cut cell mesh with fewer computational cells but a better maximum-to-minimum cell volume ratio produces a quicker and more stable DDPM simulation. Comparisons between the DDPM and EE cut cell cases showed that the two multi-phase modeling methods predict different volume fractions of solids in the system at steady state. This is most likely related to the resolution of particles in the DDPM simulations and the differences between the EE and DDPM simulation could be reduced by decreasing the number of particles per cloud in the DDPM simulations. However, this would further increase the computational time for the DDPM simulations.

Due to the difficulties in running the DDPM simulations and its stability issues, we concluded that DDPM is not worth continued use for the 1 kWe solid sorbent system at this time. Instead the research focused on modeling the reactive transport with the FLUENT® EE model and the BARRACUDA model. The BARRACUDA Eulerian–Lagrangian modeling tool is able to provide a more stable solution than DDPM but is a serial code and so expansion to larger systems could present computational issues. Additionally, simplifications were made to the geometry and operating conditions in order to reduce the complexity (number of cells) for BARRACUDA. It should be noted that with these simplifications, simulation of the full integrated system with chemical reaction was possible with BARRACUDA. The EE FLUENT® multi-phase method provided the simplest implementation and a relatively fast solution. It also required simplifications to the system and decomposed the reactor into individual components.

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References