Computational Fluid Dynamic Investigation of Change of Volumetric Flow in Fluidized-Bed Reactors

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The modeling and characterization of fluidized-bed reactors is challenging due to the variety of interrelated phenomena during the fluidization process. A key feature affecting gas–solid fluidized-bed reactors is the distribution of gas flow between phases. To address this flow distribution, conventional reactor modeling and computational fluid dynamics have been proposed in the literature. However, very little attention has been given to cases where the fluid volume changes due to factors such as variation in the total molar flow due to reaction, change of phase, utilization of membranes, and changes in temperature and pressure. This article investigates change in volumetric flow based on computational fluid dynamics. It is shown that a decrease in volumetric flow can profoundly influence the hydrodynamics, especially for smaller particles, whereas an increase in volumetric flow is accommodated with limited consequences, especially for larger particles.

1. Introduction

Fluidized-bed reactors are used in a variety of chemical and physical operations. Most reactions involve gases and particles, with the particles either acting as catalysts or participating in the reaction. However, the design, modeling, and characterization of such reactors are complex and often uncertain processes.

Modeling is an important tool for describing complex systems.1,2 Reactor models are evolving rapidly and are essential for many disciplines, including fluidization engineering. From the research point of view, reactor modeling can provide new insights for better understanding the intrinsic phenomena in fluidized-bed reactors. Fluidized-bed reactors require intricate models based on a combination of conservation equations, empirical relations, and numerous parameters. A key element in model development is balancing-interphase mass transfer.3 This term accounts for exchange of mass between the dilute and dense phases in the bed. Although this balancing term has little or no effect on systems with negligible change in molar or volumetric flow, it can play an important role in systems where the molar flow changes significantly due to reaction or other factors.

The fluid volumetric flow can be linked to changes in the total molar flow due to chemical reactions,4,5 decrease of molar flow due to gas removal through membranes, changes in temperature and/or pressure, or change of phase. For example, there is a change in the net molar flow due to the reaction in a number of reactions of practical importance. This can be an increase, as in steam methane reforming:

\[
\text{CH}_4 + 2\text{H}_2\text{O} \leftrightarrow \text{CO}_2 + 4\text{H}_2 \tag{1}
\]

where the stoichiometry is such that 3 moles on the left side gives rise to 5 on the right side. Alternatively, it can be a decrease, as in oxy-chlorination:

\[
\text{C}_2\text{H}_4 + 2\text{HCl} + \frac{1}{2}\text{O}_2 \leftrightarrow \text{C}_2\text{H}_2\text{Cl}_2 + \text{H}_2\text{O} \tag{2}
\]

where there is a stoichiometric loss of 1.5 moles per mole of ethylene reacted. Both of these reactions are at the heart of important commercial processes. Similar changes in the volumetric gas flow can occur in rapid drying and condensation in fluidized beds.

Because the vessels in which the reactions take place are opaque and very large, it is not possible to determine in a direct manner the effect of the molar flow changes in commercial reactors. This area needs further research as it is not clear how quickly additional moles are transferred from one phase to another, something that is not covered by the standard two-phase theory of fluidization.6,7 In modeling fluidized-bed reactors for processes where such volume changes take place, the modelers have had to make reasonable, but untested, assumptions about how the change in molar (and hence volumetric) flow affects the bed hydrodynamics and interphase mass transfer. At one extreme, these could, for example, lead to defluidization or great expansion of the dense phase, or, at the opposite extreme, be accommodated by immediately transferring the “extra” or “missing” flow to the other phase with negligible influence on the local hydrodynamics.

The effects of volume change are likely to be most interesting in the bubbling flow regime, but they could also play a role in the other major fluidization flow regimes (slug flow, turbulent fluidization, and fast fluidization). For simplicity, we focus here on the bubbling flow regime. In this article, volume changes in gas flow in bubbling fluidized beds caused by chemical reactions and phase change are investigated. Computational fluid dynamics (CFD) calculations are used to predict whether or not reaction-induced local volume changes profoundly affect the fluidization hydrodynamics. The predictions also give an indication of the likely interphase mass transfer implications of changes in volumetric flow occurring primarily in the dense phase.

1.1. Interphase Balancing Mass Transfer in Reactor Models. From conventional two-phase theory, it is assumed that most reaction occurs in the dense phase, whereas most flow is through the dilute phase. These factors can lead to significant...
changes in flow in the dense phase if there is no balancing flow to maintain a more appropriate flow distribution.

The effects of volume gain and, even more, volume loss could be profound, because most reaction takes place in the dense phase where the flow is small (commonly assumed to be just enough to fluidize the particles), whereas most of the gas flow is via the bubbles, which contain few particles and therefore have low reaction rates. Interphase mass transfer between the two phases therefore plays an important role. Consequently, estimating the balancing gas cross-flow is a critical step in reactor modeling when there is significant variation in the total molar flows due to reaction and/or membrane flux. To calculate the interphase crossflow between the fluidizing phases in conventional two-phase models, it is imperative to impose a flow division.

For bubbling beds, Toomey and Johnstone defined what is commonly referred to as the two-phase theory of fluidization, giving a “visible” bubble flow of:

$$v_{VB} = (U_s - U_{mf})A$$

where $$U_s$$ is the superficial gas velocity, $$U_{mf}$$ is that at minimum fluidization, and $$A$$ is the reactor cross-sectional area. There is considerable evidence that this standard two-phase theory overestimates the visible gas flow. This may be due to increased flow in the dense phase or increased throughput in the dilute phase. Grace and Clift summarized the results from experimental studies dealing with the gas flow division in fluidized beds and proposed:

$$v_{VB} = (U_s - mU_{mf})A$$

where $$m$$, based on experimental data, varied over a wide range (1.0 to ~18).

An alternative modification of the standard two-phase theory suggests:

$$v_{VB} = Y(U_s - U_{mf})A$$

where $$Y$$ is a constant less than unity, with experimental values between 0.67 and 0.8. This constant may be predicted using the dimensional correlation proposed by Peters et al., applicable only for $$U - U_{mf} \leq 0.233 \text{ m/s}$$:

$$Y = 0.7585 - 0.13(U_s - U_{mf}) + 5(U_s - U_{mf})^2$$

1.2. Strategies in the Literature Based on Conventional Two-Phase Theory Modeling. The majority of conventional fluidized-bed reactor models available in the literature ignore the effects of change in volumetric flow. Although the issue of change in volumetric flow has never been fully resolved, some earlier works treat it as follows:

Irani et al. modified the Kunii and Levenspiel model to account for volume change for single reactions and first-order kinetics. They assumed that all extra gas generated appears directly and immediately as bubbles, while maintaining the mass transfer coefficient constant.

Kai and Furusaki modified the model of Irani et al. to account for possible variations in mass transfer coefficients due to bulk flow for single reactions and first-order kinetics.

Adris fixed the volumetric flow of the higher density phase. However, he reported that the model was insensitive to changes in this value for a membrane reformer operating at low fluidization velocity with very low flux through membrane surfaces.

Abba assumed bulk transfer proportional to the difference between the high-density-phase volumetric flow and its entry value. Yet, he found little influence of this term for an oxychlorination reactor operating near the turbulent/bubbling boundary, and no effect was investigated for a reforming case study.

Constantineau et al. accounted for bulk transfer of the extra moles generated by chemical reaction. In that case, there were no membranes, and the effects of temperature and pressure were ignored. Furthermore, they assumed bulk flow to go only from the lower-density phase to the higher-density phase.

Mahecha-Botero distributed gas between the phases depending on the increase/decrease of total volumetric flow, which may change due to chemical reaction, membrane permeation, changes in temperature or pressure, or any combination of these factors. The gas flow in the dense phase was defined according to the modified two-phase theory as $$v_f = mU_{mf}$$, where $$m$$ was taken as 1.0 by default (i.e., corresponding to the standard two-phase theory). It was suggested that $$m$$ can be adjusted on the basis of the experimental measurements.

Although these methodologies implemented by conventional modelers are plausible, it is clear that they have uncertain ability to predict deviations from the standard two-phase theory. For that reason, we turn our attention to computational fluid dynamics to investigate changes in volumetric flow based on more robust techniques.

2. Numerical Models and Simulation Setup

2.1. Numerical Models. Numerical simulations were conducted on the basis of the multiphase flow with interphase exchanges (MFIX) CFD code, available from the U.S. Department of Energy’s National Energy Technology Laboratory (NETL) at http://mfix.netl.doc.gov. MFIX is a general-purpose computer code for modeling the hydrodynamics, heat transfer, and chemical reactions in fluid−solid systems. The governing equations for the solids phase based on the multifluid model are closed by kinetic granular theory, which assumes that the random motion of particles is analogous to thermal motion of molecules in a gas. A “granular temperature” proportional to the mean square of the random particle velocity based on a Maxwellian velocity distribution is defined to model the turbulent fluctuating energy of the solids phase. Constitutive relations for the solids phase stress tensor are based on kinetic theory. The hydrodynamic model equations are summarized in the Appendix. The detailed form of equations and numerical methods are described in the MFIX documentation.

2.2. Simulation Setup. In the current study, 2D simulation is employed to investigate the fundamentals of fluidization with changes in the volumetric gas flow. The numerical simulations were conducted in a domain of width 0.3 m and height 0.7 m. The particles were glass beads of diameter 485 μm, fluidized by a gas of density of 1.637 kg/m³. A uniform grid with a grid size of 5 mm was adopted in our simulations. This has been shown to be small enough to provide grid-independent solutions. The conditions for the simulations are summarized in Table 1. The simulation configuration was similar to the experimental setup of a quasi-2D column (0.015 × 0.3 × 0.7 m) in the experiments of Lavernier et al. Simulations were performed to study quantitatively the influence of volume change on the hydrodynamics.

The following boundary conditions were adopted. Constant pressure was assumed at the top boundary, so that particles were free to leave the system. For the bottom distributor, a uniform gas velocity was specified, with no particles entering or leaving.
the domain. At the lateral sidewalls, we adopted a no-slip boundary condition for the gas phase and a partial-slip boundary condition for the solid phase.25

Initially, the bed was partially filled by stationary particles to a depth of 0.30 m with a volume fraction of 0.6. The bed was then fluidized by gas passing through the bottom distributor at a superficial gas velocity of 0.24 m/s. Simulations were performed for 10 s, and results were recorded at a frequency of 100 Hz. Data for the final 5 s were analyzed. Increasing the simulation time had a negligible effect on the predicted bubble diameters.

2.3. Volume Change Due to Phase-Change (Drying and Condensation). Drying is an important application of fluidized beds. Given the widespread application of fluidized-bed dryers in the food, biotechnology, and pharmaceutical industries, extensive research on the fluid bed hydrodynamics has been conducted on drying processes. During drying, moisture is transferred from the solid phase to the drying air, leading to volume change of the gas flow. Similar changes in volumetric flow can be caused by evaporation of liquid spray into gas–solid systems, as encountered in a number of industrial applications such as spray dryers, fluid coking, and fluid catalytic cracking (FCC). Rapid evaporation of liquid droplets produces a large amount of vapor leading to substantial in situ increase in gas flow. On the other hand, the volumetric gas flow will decrease in situ if condensation takes place for the recovery of volatile materials.

In drying and condensation processes, phase-change leads to the change in the volumetric gas flow. Drying and condensation are very complex processes, with the rate of mass transfer depending on various factors, including temperature difference, moisture content, and thermodynamic properties. These processes can be simulated by assuming mass transfer between the gas and solid phases. The mass transfer rate for a single particle can be written as:

\[ q_s = k_s M_{vapor} \alpha_p (c_{bulk} - c_{surface}) \]  

(7)

where \( k_s \) is the mass transfer coefficient, \( \alpha_p = \pi d_p^2 \) is the particle (sphere) surface area, \( M_{vapor} \) is the molecular weight of vapor, \( c_{bulk} \) is the vapor concentration in the bulk phase surrounding the particles, and \( c_{surface} \) is the corresponding vapor concentration at the outer surface of the particles. The mass transfer rate per unit volume between the gas and solid phases can be derived as:

\[ q_v = \frac{6k_s M_{vapor} \alpha_p (c_{bulk} - c_{surface})}{d_p} \]  

(8)

where \( \alpha_p \) is the solid volume fraction. Equation 8 can be simply written as:

\[ q_v = k_s \alpha_p (c_{bulk} - c_{surface}) \]  

(9)

\[ \text{Table 1. Conditions for Numerical Simulations} \]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Column width, m</td>
<td>0.30</td>
<td>Column height, m</td>
<td>0.70</td>
</tr>
<tr>
<td>Pressure, Pa</td>
<td>101 300</td>
<td>Temperature, K</td>
<td>298</td>
</tr>
<tr>
<td>Gas viscosity, Pa·s</td>
<td>1.80 \times 10^{-5}</td>
<td>Gas density, kg/m³</td>
<td>1.637</td>
</tr>
<tr>
<td>Particle density, kg/m³</td>
<td>2500</td>
<td>Particle diameter, μm</td>
<td>485</td>
</tr>
<tr>
<td>Particle–particle restitution coefficient</td>
<td>0.95</td>
<td>Particle–wall restitution coefficient</td>
<td>0.80</td>
</tr>
<tr>
<td>Initial bed height, m</td>
<td>0.30</td>
<td>Initial bed voidage</td>
<td>0.40</td>
</tr>
<tr>
<td>Minimum fluidization velocity, m/s</td>
<td>0.194</td>
<td>Minimum fluidization voidage</td>
<td>0.40</td>
</tr>
</tbody>
</table>

\[ v \] Calculated from the equation of Wen and Yu.24

with \( k_v = 6k_s M_{vapor} \alpha_p \) as the volumetric mass transfer coefficient. For a batch of moist powder, drying usually involves two periods: (1) a constant-rate period and (2) a falling-rate period. However, it is beyond the scope of our study to include the complex drying kinetics in the current numerical simulations. As an initial approximation, the mass transfer rate from the solid surface area is assumed to be constant. By setting different values of \( k_v \), drying and condensation can be simulated. Table 2 summarizes the conditions for our numerical simulations of drying and condensation. At the bottom distributor, a uniform gas superficial velocity of 0.24 m/s was assumed, sufficient to fluidize the particles. Because of drying or condensation, the exit superficial gas velocities differed as listed in the table. For comparison, a base case without any interphase mass transfer was also simulated. In our simulations, properties of the solid particles are assumed to be independent of their moisture content, as during a 10 s simulation, the mass transferred between the solid and gas phases is less than 0.2% of the total solid inventory. Hence, it is reasonable to neglect changes in solid properties, such as density and particle diameter.

2.4. Volume Change Due to Chemical Reaction. To some extent, the drying and condensation mechanisms discussed above represent chemical reactions associated with phase change. However, it is difficult to take into account the change of gas density if both reactant and product are gaseous.

The ozone decomposition reaction and its reverse reaction were simulated to study the volume change resulting from chemical reactions considering the molar gain and loss in these reactions. Ozone decomposition has frequently been used to characterize fluidized-bed reactors, specifically to quantify gas–solids contact in the laboratory.26–28 This reaction requires only low concentrations of reactant, detection is rapid and accurate using fairly simple methods, and there is a measurable reaction rate at ambient temperature and pressure. The simple reaction network and kinetics, as well as the large amount of published experimental data, make it convenient for CFD model evaluation.29–31 However, experiments have always been carried out at very low ozone concentrations, too low for appreciable variations in volumetric gas flow due to reaction. Therefore, we assume much higher concentrations for the purpose of qualitative study.

The following assumptions were used in the simulations. Both the ozone decomposition and the reverse reaction were assumed
Table 3. Conditions for Numerical Simulations of Cases with and without Chemical Reaction

<table>
<thead>
<tr>
<th>parameters/cases</th>
<th>$O_3 \rightarrow O_2$</th>
<th>no reaction</th>
<th>$O_2 \rightarrow O_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>gas mixture</td>
<td>1:1</td>
<td>1:1</td>
<td>1:1</td>
</tr>
<tr>
<td>$(O_3:O_2)$ molar ratio</td>
<td>1.57</td>
<td>0</td>
<td>2.36</td>
</tr>
<tr>
<td>reaction rate constant, $k_r$, 1/s</td>
<td>0.24</td>
<td>0.24</td>
<td>0.24</td>
</tr>
<tr>
<td>superficial gas velocity at distributor, $U_{g,in}$, m/s</td>
<td>0.277</td>
<td>0.24</td>
<td>0.208</td>
</tr>
<tr>
<td>superficial gas velocity at top exit, $U_{g,exit}$, m/s</td>
<td>0.083</td>
<td>0.046</td>
<td>0.014</td>
</tr>
</tbody>
</table>

The reaction rate for ozone in the gas phase can be calculated as

$$R_{O_3} = -\frac{k_f \alpha_p \rho_x X_{O_3}}{M_{O_3}}$$ (11)

where $k_f$ is the first-order reaction rate constant, $X_{O_3}$ is the mass fraction of ozone in the gas mixture, and $M_{O_3}$ is the molecular weight of ozone. Because the decomposition depends on the presence of catalyst, the solid volume fraction, $\alpha_p$, is included in the kinetic rate.

For the purpose of simulation and comparison, we also considered the reverse reaction:

$$3O_2 \rightarrow 2O_3$$ (10)

with a reaction rate of

$$R_{O_2} = -\frac{k_r \alpha_p \rho_x X_{O_3}}{M_{O_3}}$$ (13)

Our simulations followed the procedure described by Syamlal and O’Brien.29 Mixtures of ozone and oxygen (composition 1:1) were assumed to be fed from the bottom distributor at a constant flow rate. Ozone decomposition and ozonation rate constants were assumed, with conditions for numerical simulations summarized in Table 3.

3. Results and Discussion

3.1. Drying and Condensation. Figure 1 shows snapshots of voidage contours for condensation and drying in comparison with the base case at $t = 7.5$ s. Differences can be clearly observed from these plots with respect to bubble sizes. In the condensation case, the bubble size is much smaller than for the base case as the gas flow rate decreases due to condensation. The bed surface is almost flat as the very small bubbles breaking the surface cause only small fluctuations. The bubble size barely increases as they rise, even decreasing in some cases. In addition, there are more bubbles than for the base case in the middle of the bed due to evaporation from the solid surfaces. However, the bubble size for the condensation case is almost constant at different levels, decreasing near the bed surface as the gas in the bubbles diffuses into the emulsion phase and then condenses. It can also be observed from the figure that the volume-change of gas flow in the dense phase affects the formation of bubbles above the bottom distributor. Bubbles tend to be detected at lower levels for the drying cases than for the condensation case.

3.2. Ozone Decomposition. Similar results were obtained for cases with chemical reaction. With a decrease in volumetric flow for the ozone generation reaction, $3O_2 \rightarrow 2O_3$, the bed expansion decreases slightly, and bubbles are predicted to be smaller than for the case where the volumetric flow increases.

3.3. Interphase Mass Transfer. As the volumetric flow in the dense phase changes, the interphase mass transfer between bubbles and the emulsion phase plays an important role. Given fast enough interphase mass transfer, all excess gas in the emulsion phase due to drying or volume-increasing chemical reactions is quickly transferred convectively to the bubbles, whereas for condensation or volume-decreasing chemical reactions, loss in gas flow in the dense phase is rapidly replaced.
from the dilute phase. The numerical results demonstrate the situation when the interphase mass transfer rate is high. For the cases investigated, the ratio between bubble velocity, $u_b$, and the emulsion gas velocity, $u_f = U_{mf}/\varepsilon_{mf}$, is less than 1, leading to cloudless (“slow”) bubbles, with bubbles rising more slowly than the gas in the dense phase. Gas in the dense phase then enters the bottom of the bubble and leaves at the top, with the bubbles providing a shortcut for faster-moving percolating gas. For cloudless bubbles, the resistance to mass transfer from the bubbles to the dense phase is small, so gas in both phases tends to have the same composition.

Figure 1. Snapshots of voidage contour at $t = 7.5$ s for (a) condensation, (b) base case, and (c) drying. For simulation conditions, see Table 2.

Figure 2. Variation of mean bubble diameter with height for the base case (error bar: ± one standard deviation). For simulation conditions, see Table 2.

Figure 3. Variation of the average bubble shape factor with height for the base case (error bar: ± one standard deviation). For simulation conditions, see Table 2.

Figure 4. Variation of mean bubble diameter with height for the base case as compared to cases where there is condensation and drying. For simulation conditions, see Table 2.

Figure 5. Variation of the average bubble shape factor with height for base case as compared to cases where there is condensation and drying. For simulation conditions, see Table 2.
undergo continuous deformation, are unlikely to be accurate frictional models, heavily based on the assumption that particles the surface friction between individual particles. Available predicted that gas would be sucked in from the top outlet. In fact, for such a high condensation rate, it is defluidizes. In such circumstances, the gas properties, such as density and viscosity. However, in gas reactions, the volume change is accompanied by the change in gas density and viscosity as the reaction takes place. For this reason, the ozone decomposition reaction was simulated to investigate the influence of simultaneous change in both volumetric flow and density on the flow hydrodynamics. According to the two-phase theory, the dense phase is subject to minimum fluidization conditions, while the excess gas rises in the form of bubbles. In the emulsion phase, Ergun’s equation suggests that

\[
(1 - \epsilon_{mf})(\rho_p - \rho_g)g = 150 \frac{(1 - \epsilon_{mf})^2 \mu_g U_{mf}}{\epsilon_{mf}^2} \frac{(\phi d_p)^2}{\mu d} + 1.75 \frac{(1 - \epsilon_{mf}) \rho_g U_{mf}^2}{\epsilon_{mf}^3} \frac{1}{\phi d_p}
\]

where \(\epsilon_{mf}\) is the minimum fluidization voidage, and \(U_{mf}\) is the minimum fluidization velocity; \(\mu_g\) is the gas viscosity, and \(\rho_p\) and \(\rho_g\) are densities of particle and gas, respectively; \(d_p\) is the particle diameter and \(\phi\) is the particle sphericity, which is unity for spherical particles. In eq 14, the first term on the right side is due to viscous drag and is the dominant term in the laminar flow regime; the second term results from the inertial effects and is dominant in the turbulent flow regime. Provided that \(\epsilon_{mf}\) is constant and \(\rho_p \gg \rho_g\), \(U_{mf}\) decreases as \(\rho_g\) increases, a prominent trend for flow in the turbulent regime. This is consistent with previous work on effect of pressure, as investigated by many researchers. Rowe reported that \(U_{mf}\) was nearly independent of pressure for small particles \((d_p < 100 \mu m)\), but decreased sharply with increasing pressure for large particles \((d_p > 500 \mu m)\). Consequently, it might be inferred that coarse particles would remain fluidized when the gas volume decreases, but the density increases due to reaction. However, smaller particles may defluidize under the same circumstance.

To confirm this finding, particles of diameter 200 \(\mu m\) were simulated for ozone decomposition and ozonation with a fine grid of size 2.5 mm for flow conditions summarized in Table 4. For these particles, the ratio between bubble velocity, \(U_b\), and the emulsion gas velocity, \(u_e\), is larger than 1, leading to “fast” bubbles with clouds, with high reactant concentration confined to the cloud and limited interphase mass transfer between the bubbles and dense phase.

The mean bubble size variations along the height are compared in Figure 9 for different cases. There is very little change in mean bubble sizes for all cases, although the volumetric flow rate changed due to reactions. A slight decrease in bubble size is observed for both ozone decomposition and ozonation. The slight decrease for the decomposition case is caused by the formation of small bubbles in the upper region. Because of the limited interphase mass transfer between bubbles and the emulsion phase, the excess gas produced by the volume gain reaction is predicted to nucleate new bubbles. For the ozonation case, the decrease in bubble size is small because of
the relatively small change in volumetric flow and the low interphase mass transfer. The interphase mass transfer rate for fine particles is smaller than for the coarse particles due to the reduced throughflow velocity and cloud formation. This slower interphase mass transfer reduces the rate at which bubble size adjusts to the changes in volumetric flow.

Table 4. Conditions for Numerical Simulations of Beds with Fine Particles

<table>
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<th>parameters/cases</th>
<th>O₃ → O₂</th>
<th>no reaction</th>
<th>O₂ → O₃</th>
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<tbody>
<tr>
<td>gas mixture</td>
<td>1:1</td>
<td>1:1</td>
<td>1:1</td>
</tr>
<tr>
<td>(O₃:O₂) molar ratio</td>
<td></td>
<td></td>
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</tr>
<tr>
<td>reaction rate</td>
<td>1.57</td>
<td>0</td>
<td>2.355</td>
</tr>
<tr>
<td>constant, kₑ, 1/s</td>
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</tr>
<tr>
<td>superficial gas velocity at distributor, Uₑ,m, m/s</td>
<td>0.09</td>
<td>0.09</td>
<td>0.09</td>
</tr>
<tr>
<td>superficial gas velocity at top exit, Uₑ,exit, m/s</td>
<td>0.112</td>
<td>0.09</td>
<td>0.077</td>
</tr>
<tr>
<td>Uₑ,exit - Uₑ,exit, m/s</td>
<td>0.074</td>
<td>0.052</td>
<td>0.039</td>
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the relatively small change in volumetric flow and the low interphase mass transfer. The interphase mass transfer rate for fine particles is smaller than for the coarse particles due to the reduced throughflow velocity and cloud formation. This slower interphase mass transfer reduces the rate at which bubble size adjusts to the changes in volumetric flow.
Predicted pressure drops across the bed are plotted for both types of particles in Figure 10. The pressure drop corresponding to the bed weight minus buoyancy, $\Delta P = (\rho_b - \rho_f)(1 - \epsilon_0)\rho_b g h_0$, is also plotted. For both types of particles, the standard deviation of pressure drop increases with increasing exit excess superficial gas velocity, indicating larger bubbles erupting at the bed surface. For coarse particles, due to the fast interphase mass transfer, the mean pressure drop is very close to the theoretical value (bed weight minus buoyancy divided by cross-sectional area), indicating that the bed is well fluidized. For finer particles, the pressure drop for the ozonation reaction deviates appreciably from the theoretical value, presumably associated with local defluidization of the dense phase. Experimental verification of these results is needed. An experimental study on the change of volumetric flow resulting from sudden increase/decrease in pressure is in progress and will be reported elsewhere.\textsuperscript{41}

4. Conclusions

The influence of volume change of gas phase due to phase-change and chemical reactions in the fluidized beds was studied numerically with a Eulerian–Eulerian CFD model. For changes in volumetric flow, bubble size and shape factor were analyzed. For coarse particles with fast interphase mass transfer between bubbles and the dense phase, bubble size increases or decreases due to gas volume changes in the dense phase. Even with substantial volume loss, no defluidization is predicted provided that the overall superficial gas velocity always exceeds the minimum fluidization velocity. However, for finer particles, changes in bubbles sizes are slow to adjust when the volume increases or decreases in the dense phase, and interphase mass transfer plays an important role. Defluidization could then take place, even when the overall superficial gas velocity is much higher than the minimum fluidization velocity for cases where there is \textit{in situ} loss of volumetric gas flow due to reaction or condensation.

Appendix: Summary of MFIX Equations

Governing Equations.

a. Continuity Equations.

$$\frac{\partial}{\partial t}(\alpha_g \rho_g) + \nabla \cdot (\alpha_g \rho_g \vec{V}_g) = R_{gp} - R_{pg}$$

b. Momentum Equations.

$$\frac{\partial}{\partial t}(\alpha_g \rho_g \vec{V}_g) + \nabla \cdot (\alpha_g \rho_g \vec{V}_g \vec{V}_g) = -\nabla p + \alpha_g \Sigma p + \alpha_p \rho_p \vec{V}_p + R_{pg} \vec{V}_g - R_{gp} \vec{V}_p + I_{gp}$$

Constitutive Equations.

a. Gas Stress Tensor.

$$\bar{\tau}_g = 2\mu_{ge} \bar{S}_g$$

$$\bar{S}_g = \frac{1}{2} \nabla \vec{V}_g + (\nabla \vec{V}_g)^T - \frac{1}{3} \nabla \cdot \vec{V}_g \bar{I}$$

$$\mu_{ge} = \min(\mu_{max}, \mu_g + \mu_c)$$

$$\mu_c = 2\bar{\rho}_g \rho_g \sqrt{T_{Dg}}$$

$$\bar{D}_g = \frac{1}{2} (\nabla \vec{V}_g + (\nabla \vec{V}_g)^T)$$

b. Solids Stress Tensor.

$$\bar{\tau}_p = -P_s + \eta \mu_{gs} \nabla \cdot \vec{V}_p \bar{I} + 2\mu_{sp} \bar{S}_p$$

$$\bar{S}_p = \frac{1}{2} (\nabla \vec{V}_p + (\nabla \vec{V}_p)^T) - \frac{1}{3} \nabla \cdot \vec{V}_p \bar{I}$$

$$P_s = \alpha_p \rho_p \Theta \left[1 + 4g_0(\alpha_p)\right]$$

$$\mu_p = \left(\frac{2 + \alpha}{3}\right) \frac{\mu_p^*}{g_0(2 - \eta)} \left(1 + \frac{8}{3} \eta g_0(\alpha_p) \times \left(1 + \frac{8}{3} \eta(3\eta - 2)g_0(\alpha_p) + \frac{8}{3} \eta \mu_0\right)\right)$$

$$\mu_p = \left(\frac{2 + \alpha}{3}\right) \frac{\mu_p^*}{g_0(2 - \eta)} \left(1 + \frac{8}{3} \eta g_0(\alpha_p) \times \left(1 + \frac{8}{3} \eta(3\eta - 2)g_0(\alpha_p) + \frac{8}{3} \eta \mu_0\right)\right)$$

$$\frac{\partial}{\partial t}(\alpha_p \rho_p \vec{V}_p) = R_{pg} - R_{gp}$$

Figure 10. Pressure drop across the bed for (a) coarse particles and (b) finer particles. For simulation conditions, see Table 4.
\[ \mu_p = \frac{\alpha_p \rho_p \Theta_p g_0 \mu}{\alpha_p \rho_p \Theta_p g_0 + \frac{2 \mu}{\alpha_p \rho_p}} \]
\[ \mu = \frac{5}{96} \rho_p d_p \frac{\gamma \pi \Theta_p}{\eta} \]
\[ \mu_b = \frac{256}{5 \pi} \mu a_c^2 \eta \]
\[ \eta = \frac{1 + e}{2} \]

\section*{c. Granular Temperature.}
\[ \frac{3}{2} \frac{d}{dt} (\alpha_p \rho_p \Theta_p) + \nabla \cdot (\alpha_p \rho_p \nabla \Theta_p) = \frac{7}{3} \frac{\mu}{\alpha_p} \nabla \cdot (\frac{\nabla \Theta_p}{\alpha_p \rho_p}) - \alpha_p \rho_p \beta \]
\[ \kappa_{p,\Theta} = \frac{\kappa^*}{g_0} \left(1 + \frac{12}{5} \eta \alpha_p \gamma \right) \left(1 + \frac{12}{5} \eta^2 (4 \eta - 3) \alpha_p \gamma^2\right) + \frac{64}{25 \pi} (41 - 33 \eta) \alpha_p \gamma^2 \]
\[ \kappa_p = \frac{\rho_p \alpha_p \gamma \Theta_p \kappa}{\rho_p \alpha_p \gamma \Theta_p + \frac{64 \kappa}{5 \rho_p \alpha_p}} \]
\[ \kappa = \frac{75 \rho_p d_p \frac{\gamma \pi \Theta_p}{\eta}}{48 \eta (41 - 33 \eta)} \]

\section*{d. Interphase Momentum Exchange.}
\[ I_{bg} = \beta (V - \bar{V}) \]
\[ \beta = \left\{ \begin{array}{ll}
150 \frac{\alpha_p \rho_p}{\alpha_p \rho_p} \frac{|V - \bar{V}|}{d_p} & \text{if } \alpha_p > 0.2 \\
1.75 \frac{\alpha_p \rho_p}{\alpha_p \rho_p} \frac{|V - \bar{V}|}{d_p} & \text{if } \alpha_p \leq 0.2 \\
3 \frac{C_d \alpha_p}{d_p} \frac{|V - \bar{V}|}{d_p} & \text{if } \alpha_p < 0 \end{array} \right. \]
\[ C_d = \frac{24}{Re \cdot \alpha_p} (1 + 0.15 (Re \cdot \alpha_p)^{0.687}) \text{ if } Re \cdot \alpha_p < 1000 \\
0.44 \text{ if } Re \cdot \alpha_p \geq 1000 \]
\[ Re = \frac{\rho_p (|V - \bar{V}| d_p)}{\mu g} \]

\section*{Greek Symbols}
\[ \alpha_d \] gas volume fraction
\[ \alpha_p \] solid volume fraction
\[ e_0 \] initial bed voidage
\[ \epsilon_{mf} \] minimum fluidization voidage
\[ \epsilon \] particle sphericity
\[ \mu_g \] gas viscosity, Pa \cdot s
\[ \rho_g \] gas density, kg/m^3
\[ \rho_p \] particle density, kg/m^3

\section*{Literature Cited}
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