REAL-TIME BUBBLE SIMULATIONS FOR FLUIDIZED BEDS

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INTRODUCTION

Fluidized bed reactors are widely used in the chemical industry and are essential to the production of key commodity and specialty chemicals such as petroleum, polymers, and pigments. Fluidized beds are also going to be widely used in the next generation power plants in aiding conversion of coal to clean gas. However, in spite of their ubiquitous application, understanding of the complex multi-phase flows involved is still very limited. In particular, existing computer simulations are not sufficiently accurate/fast to serve as a primary approach to the design, optimization, and control of industrial-scale fluidized bed reactors. Availability of more sophisticated computer models is expected to result in greatly increased performance and reduced costs associated with fluidized bed implementation and operation. Such improved performance would positively affect U.S. chemical/energy industry competitiveness and increase energy efficiency.

To improve fluidization simulation capabilities, two different projects are undertaken at ORNL with the specific objective of developing improved fluidization computer models. On one hand, a very detailed multiphase computer model (MFIX) is being employed. On the other hand the Dynamic Interacting Bubble Simulation (DIBS) is being further developed at ORNL with the eventual aim of real time diagnosis and control of industrial scale fluidized beds.

The details of the DIBS model are reported in recent papers/presentations [1–5]. Here only the salient results are reported.

RESULTS AND DISCUSSION

For understanding and evaluating the predictions of our model, we compare it to experimental measurements of ozone decomposition in a bubbling bed made by Fryer and Potter [6]. The bed schematic is shown in Fig. 1. Because the observations of Fryer and Potter were time-averaged, we compute corresponding averages from our model output. Later, we discuss the dynamical features in our model output that were not measured by Fryer and Potter.

The first comparison we make with experiment focuses on the time-average bed hydrodynamics; specifically, bubble size and bed expansion. Next we consider time-average chemical conversion.

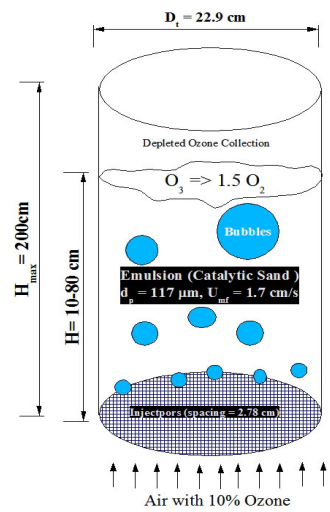

Fig. 1. Configuration of the ozone decomposition experiment—time-average comparisons.

BUBBLE SIZE

The performance of fluidized beds is usually dominated by the bed hydrodynamics. Traditionally, the time-average hydrodynamic features of greatest interest have been the axial variation of bubble size and overall the bed expansion as functions of gas flow. Figure 2 compares our predictions of bubble size versus bed height at one flow with the observations of Fryer and Potter and the widely used correlations of Darton et al. [7] and Werther [8]. In their experiments, Fryer and Potter measured bubble diameter at different heights by physically changing the amount of bed material and observing the bubble eruptions at the surface. In doing this, they assumed that such measurements were equivalent to the bubble sizes at corresponding locations in a deep bed. We note that the sizes predicted by the correlations consistently fall below those reported by Fryer and Potter.

Also plotted on the figure are the results from the model. The bubble diameter at the injectors is prescribed based on the experimental correlation as a function of excess superficial velocity as obtained by Fryer and Potter. The red line (with symbol ‘x’) is the variation of the bubble size (number mean) with axial distance for a bed of height 80 cm. The initial part of the variation (up to 40 cm) matches quite well with the experiments. The bubble diameter in the later part is under predicted. In order to address the assumption mentioned above, we simulated the experimental procedure with varying bed heights and observed the eruption diameter. From the plot it is apparent that the actual bubble diameter and the eruption diameter are in agreement for shallow beds but are different as we traverse taller beds. The eruption diameter as predicted by the model is closer to the experiments than the actual bubble diameter

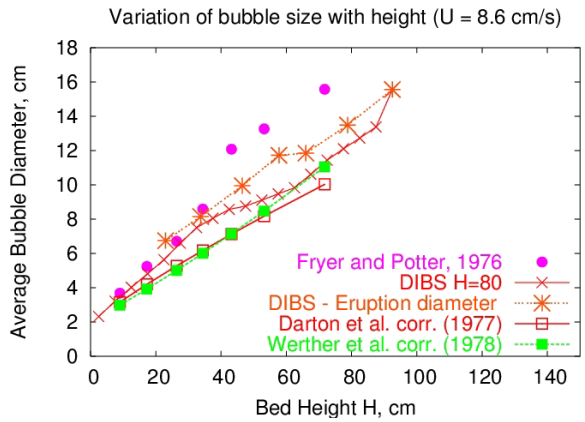

Fig. 2. Bubble-size variation with axial distance and bed height.

in a deep bed. Considering the simplicity of the model, this is very good agreement. In the next section, we point out considerable differences between various measures of the bubble size. This ambiguity points to the need to carefully evaluate the potential weighting (i.e., biasing effects) of the experimental measurement approach.

In Fig. 3, bed expansion is plotted as a function of the inlet gas velocity. As gas velocity increases, the volume of the bubbles increases and leads to greater bed expansion. Again our bubble model appears to predict the expansion with reasonable accuracy. This gives us some confidence that at least the time average aspects of our model are not too far from reality.

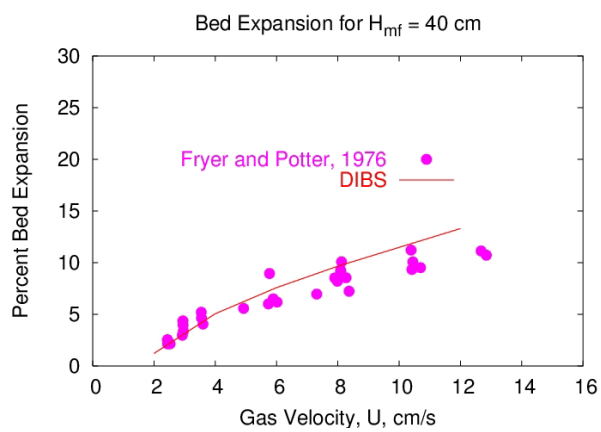

Fig. 3. Bed expansion versus gas velocity.

Other important time-average quantities that were not available here were the bubble velocities and the bubble-size distributions. Perhaps measurement of these quantities should be a future experimental goal for ozone reactors. The bubble-size distributions predicted by our model (Fig. 4) have a non-

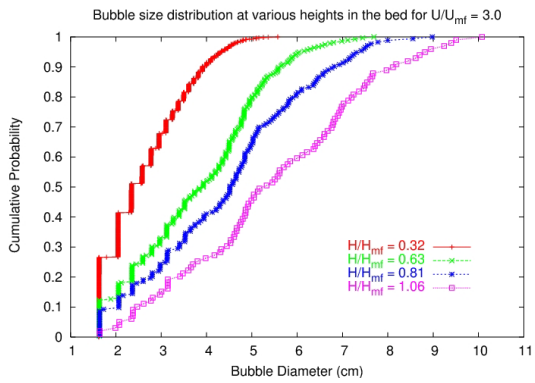

Fig. 4. Variation of bubble size distribution along the axis of the bed—ozone conversion.

Gaussian power law form that appears to scale in a self-similar fashion as one moves upward in the bed. Such scaling may be useful for diagnostics and correlations and may have some general implications for modeling. Some of these ideas have already been suggested by Gheorghiu et al. [9]. We plan to investigate these size-distribution effects further with our model in the future.

The ozone concentration at the exit is plotted for different catalyst activity (different intrinsic reaction rates) and for different gas velocities in Fig. 5. Conversion becomes reaction-rate limited at the lower reaction rates, while it becomes mass-transfer limited at higher reaction rates. The results shown here are using the model with a constant mass-transfer correction factor of 2.0 for all the cases. The model output for all the cases is in reasonably good agreement with the experiments for all the cases. Slight over prediction of ozone concentration for $k = 7.75\text{s}^{-1}$ for high gas velocities where it is mass-transfer limited is not very surprising considering all the assumptions made in the simulations. In the current model, we do not account for reactions in the wake of the bubble, and that can easily accommodated using known correlations. In addition, the emulsion-phase circulation is not currently modeled, and thus effects of solids back-mixing are not captured. The recirculation of the solids can increase the effective emulsion-gas residence time and thus increase conversion. Fryer and Potter suggested the minimum they observed in axial ozone concentration might be due to this effect. In the future, we plan to introduce both internal bubble reactions and emulsion-phase circulation to the model.

In the above discussion, we compare time-averaged statistics of the model with those of the experiments. The temporal patterns are lost in such comparisons, so we now consider the dynamical predictions of our model.

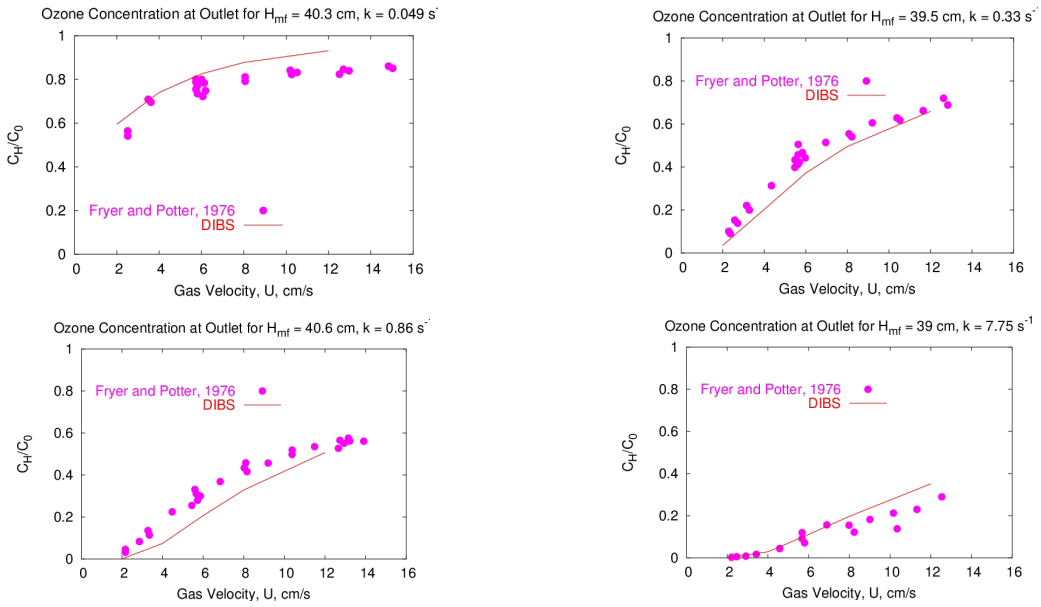

Fig. 5. Ozone conversion for various reaction rates and inlet gas velocities—temporal predictions.

FORMATION OF CHANNELS (GAS BYPASS)

A characteristic feature of our model and experimental bubbling beds is a tendency for the bubbles to collect into a persistent central channel. In the model, this channeling behavior is a natural consequence of the bubble interaction rules and the geometric boundary conditions. Specifically, given an initially uniform dispersion of bubbles at the distributor, it is always more likely that a trailing bubble will be caught in the influence of a leading bubble closer to the bed center than one further from the center. Once coalescences begin, the bias towards the center becomes further skewed because the coalesced bubbles are already displaced in that direction. The global effect of these sequential coalescences is to create an avalanche of inward moving bubble gas. Thus it is not necessary for there to be some external lateral force introduced near the walls (e.g., associated with downward moving solids) to achieve channeling. One could argue that the observed downward solids circulation near the walls is a natural result of the concentration of bubbles near the center. The avalanche of coalescences is also responsible for the overall pulsation in gas flow.

Figure 6 illustrates a simulation that resulted in the formation of a central bubble channel in a 3-D bed. The figure shows snapshots of the bubbles at different times of the simulation. This case corresponds to gas velocity of 8.6 cm/s with k = 7.75 s^{-1} in Fig. 5. The snapshots give indication of bubble size distribution and also the ozone conversion as indicated by the color. Blue corresponds to the inlet concentration of ozone, while red corresponds to almost full conversion. There are several things that can be noted from these pictures. First there is period of rapid convergence where big bubbles are formed and bubble gas passes rapidly through the middle of the bed. This is followed by period in which there are

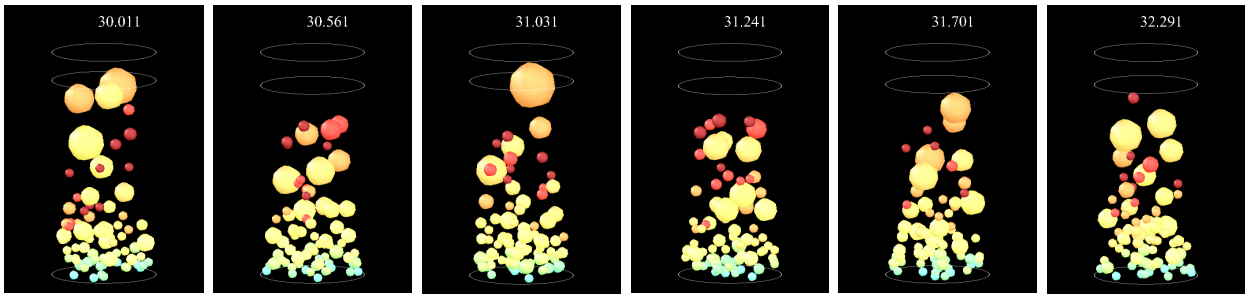

Fig. 6. Channel Formation in a Bubbling Bed .

many smaller bubbles moving upward more slowly. As the smaller bubbles become more and more numerous, a critical 'avalanche' condition is reached that triggers another round of rapid coalescence. The net result of these alternating processes is that the bubble gas pulses through a central channel that is continually reforming. This phenomenon is more pronounced in deeper beds (Halow et al., 1998; Pannala et al., 2001, 2003) where H_{mf}/D_t is larger than 5.

The pulsing flow also affects conversion. The injected bubbles have high concentrations of ozone and if the bubble size remains small, there is a good surface-to-volume ratio to ensure proper mass transfer between the bubbles and emulsion and thus resulting in higher conversion of ozone. The coalescence of the bubbles leads to bigger, faster moving bubbles with lower surface-to-volume ratio and thus exhibit poor conversion. Understanding these temporal oscillations might make it possible to perturb the bed in ways that counteract a direction to minimize the coalescence of the smaller bubbles into bigger bubbles and thus improve the conversion.

BED DYNAMICS

Figure 7 shows the number of bubbles with time for the above simulation. The simulations are run for 90 seconds, and the statistics are collected over the last 60 seconds of the simulation. Here we plot a 20 second window to illustrate the complex temporal variations. Figure 8 plots the time trace of the bed height for the same 20-second period. Bed height also behaves similarly to bubble number. For the two-phase model, one expects bed height to be directly proportional to the volume of gas in the bubble phase. From the plot it is apparent that there is are characteristic oscillations with periods of around 1 and 4 seconds.

Figure 9 illustrates the effect of the type of bubbles-size averaging on the observed trends. Average bubble diameters are computed based on number, area, volume and volume-to-surface ratio (Sauter mean or SMD). The number mean seems to be the smoothest and SMD seems to have the maximum variations. These differences indicate that the type of measurements made (i.e., the type of weighting involved in sampling) can affect the visibility of the dynamics. For evaluating gas bypassing through the bubbles, it

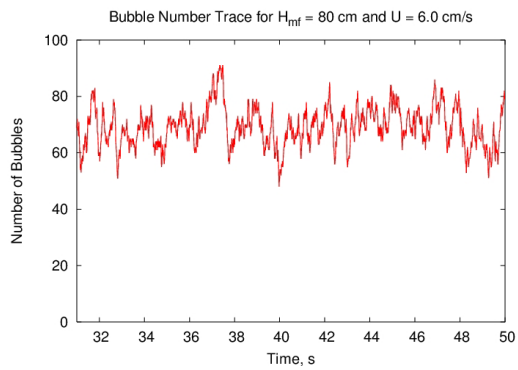

Fig. 7. Time trace for number of bubbles.

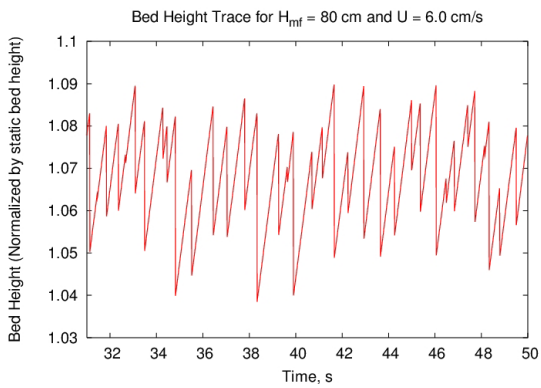

Fig. 8. The bed-height variation exhibits chaotic behavior.

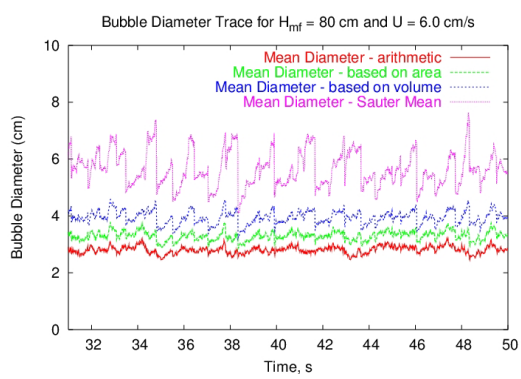

Fig. 9. Time trace of bubble size [diameter based on number mean, area, volume and Sauter mean diameter (3,2)].

would appear that SMD is more sensitive to the bubble dynamics. One also expects that the SMD bubble size would correlate most directly with conversion. Figure 10 illustrates this in terms of the average ozone concentration (normalized by the inlet concentration) in the bed overall and in the bubble phase as functions of time. Note that as the catalytic activity is increased, the relative variations in ozone

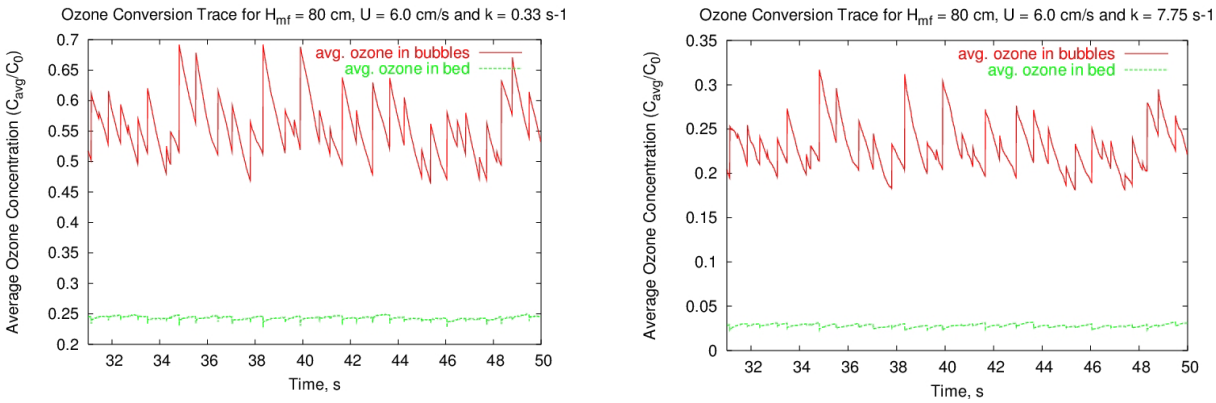

Fig. 10. Variation of Ozone concentration with time.

concentration become even more pronounced, and the ozone concentration is much lower in both measures.

In order to better quantify the temporal patterns, we computed auto-correlations, cross-correlations and power spectral density for some of the computed quantities. Figure 11(a) is a plot of the auto-correlation function for bed-averaged ozone concentration, bed height, bubble number, and SMD. Here we observe strong oscillations in all these quantities over time scales of a few seconds to tens of seconds. Figure 11(b) is a plot of cross correlations between bed-average ozone concentration and bed height, number mean bubble diameter and SMD. It is clear that ozone concentration in the bubble phase is strongly correlated with SMD. As expected, the cross correlation at zero time lag between ozone concentration in the bubbles and SMD is strongly negative, reflecting the impact of bubble size on mass transfer. The effect of bed pulsation is evident at longer time scales where bubble ozone concentration and bubble size become positively correlated. On the contrary, the overall average ozone concentration in the bed has a positive correlation with bubble size at small times that is out of phase with the bubble gas. We speculate that this difference in phase is due to the time delay associated with mass transfer and might potentially be used as a way to indirectly measure mass transfer. At further time delays, the cross correlations between gas phase and bubble phase ozone and bubble size appear to continue to shift in and out of phase. Figure 11(c) illustrates the cross-correlation between bed height with the number of bubbles and volume average bubble diameter. It is evident from this plot that bed height and volume average bubble diameter are strongly correlated, but there is an unexpected anti-correlation with the number of bubbles.

Fourier power spectra are useful for further resolving some of the oscillation time scales. As shown in Fig. 11(d–e), the power spectral densities of the bed ozone concentration, bed height, number of bubbles, and SMD reveal strong oscillations below 1 Hz. The latter three quantities have similar periods, but the ozone concentration is definitely varying at a lower frequency (about half) compared to the others.

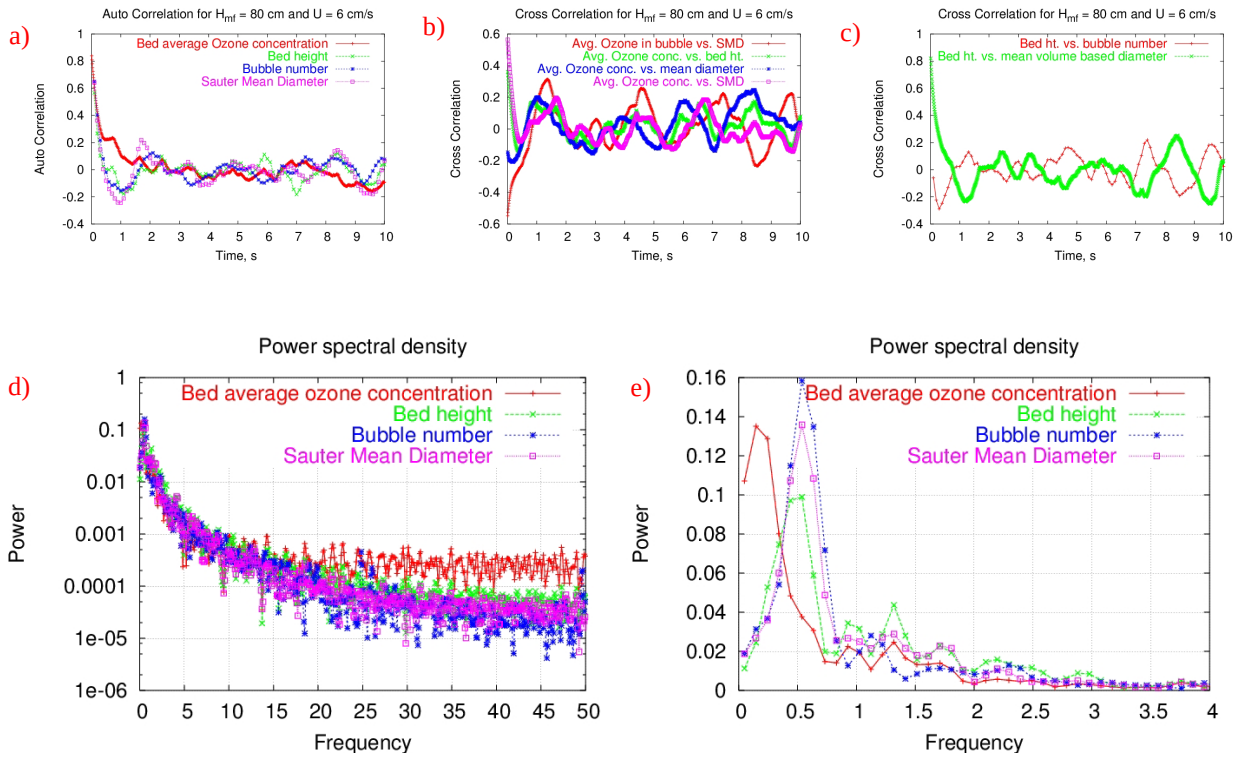

Fig. 11. Time series analysis of the model output: (a) Auto-correlation, (b) Cross-correlation for ozone concentration (c) Cross-correlation for bed height, and (d–e) Power spectral density.

LONG TIME SCALE FEATURES

In the above discussion, we observe that many dynamical features occur over time scales of seconds to tens of seconds. We observe from our model, however, that there are some global features that occur over even longer time scales. Because our model runs so fast, it is possible to explore these features in relatively great detail. Long time scale dynamics are reported in Daw et al. (2003).

SUMMARY

We find that a simple model of bubble dynamics combined with appropriate mass transfer and chemistry can simulate many key features of bubbling bed reactors. In addition, such a model predicts important spatial and temporal variations in conversion and performance that need to be further investigated. Specifically, we observe large variations in the degree of gas bypassing that might be altered by making appropriately timed external perturbations. These large variations occur as the result of the formation of bubble chains that grow and then catastrophically coalesce. We conjecture that simple models such as ours may provide a practical way for implementing model-based control of these global processes.

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