Perspective

Modeling the Hydrodynamics of Multiphase Flow Reactors: Current Status and Challenges

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Few things are more central to chemical engineering than multiphase flow chemical reactors; they are used in industry to produce a variety of chemicals, where economy of scale remains the driving factor. The engineering issues are classical, and the modern trend towards miniaturization will have little impact on these systems.

Multiphase reactors for classic applications such as fluid catalytic cracking are still evolving. New processes such as slurry bubble column reactors for gas conversion are under development. Reliable multiphase reactor models that can be used with confidence for improving existing processes and scale-up of new processes are not yet available.

Good contacting between phases in multiphase reactors is essential to promote interphase transport of species and energy. However, in many instances this is hard to achieve, as the state of uniform spatial distribution of the various phases is unstable and gives way to nonuniform structures spanning a wide range of length and time scales. Macro-scale coherent structures, which are responsible for large-scale mixing, are commonly observed (van den Akker, 1998). Their formation, growth, and propagation are influenced by reactor size, and inlet and exit configurations; this relationship is poorly understood. Consequently, we continue to rely on expensive pilot-scale cold-flow experiments and remain unsure of the effects of heat transfer, operating pressure, and temperature (Yates, 1996; Al-Dahhan et al., 1997; Fan et al., 1999), and reactions on the flow behavior.

Technology Vision 2020: The Chemical Industry—a report published in 1996 by five organizations representing the chemical industry, including AIChE—has identified a better understanding of gas-solid and gas-liquid-solid flows in chemical reactors as a critical need. It calls for the development of enabling technologies in the form of reliable simulation tools that integrate detailed models of reaction chemistry and computational fluid dynamic (CFD) modeling of macro-scale flow structures.

The most practical approach to simulating the hydrodynamics of commercial-scale multiphase reactors is through continuum models that treat the coexisting phases as interpenetrating continua. The general structure of the continuity and momentum balance equations is the same for all dispersed two-phase flow problems, although the closure relations are system-dependent. This is rather convenient, as one can use a single general-purpose multiphase CFD code for all the systems. Even more interestingly, there is a remarkable similarity in the hierarchy through which nonuniform solutions emerge out of the state of uniform multiphase flow in seemingly different systems such as fluidized beds and trickle beds. There is ample experimental evidence to suggest that these similarities are real (Krishna et al., 1998).

Coarse-grid simulation of macro-scale flow structures

Macro-scale coherent structures in multiphase flow, particularly those involving vortical motion, can be captured only through three-dimensional (3-D) transient simulations. A 3-D simulation of two-phase flow using, say, a million nodes, involves the solution of over nine million nonlinear equations at each time step; the size of the problem becomes much larger when energy and species balances are included. Simulations of such magnitude are not common in process design today, but will become routine in the not-too-distant future. As we shall see, even with this many nodes, the grid structure in simulations of commercial-scale reactors remains coarse; so, the model equations used to simulate commercial reactor performance must represent averages at this coarse-grid scale. The drag and stresses influencing the hydrodynamics and effective dispersion coefficients and reaction rates appearing in the energy and species balance equations are affected significantly by the subgrid scale fluctuations and are grid-size dependent. Yet, this dependence is just beginning to be appreciated. Experiments, both laboratory and computational, and theory that will lead to accurate scale-dependent closures for these quantities represent important frontiers in this class of problems.

The coarse-grid simulation of multiphase flow is conceptually similar to large eddy simulation of single-phase turbulent flow, where one accounts for the effects of unresolved eddies through sub-grid models (Fox, 1996). In single-phase turbulent flow, the flow of energy associated with fluctuations is predominantly from large scale to smaller ones. In the class of multiphase flows considered here, meso-scale structures arise as a result of local instabilities and grow into larger and larger scales, so macro-scale shear is not a requirement for creating and sustaining a chaotic state of flow (unlike in single-phase flow). Because of this difference, we cannot simply adopt the ideas developed in single-phase flow. In this article, we examine what is known generically about flow structures at different scales in gas-solid, gas-liquid, and gas-liquid-solid systems, and point out where challenges lie in building the hydrodynamic components of the next generation of reactor models.

Gas-solid flows

Industrial processes involving fluidized gas-solid suspensions typically employ particles having a wide-size distribution with an average diameter in the range of 50 to 100 μm. The volume fraction of particles range from as low as 0.01 in riser tubes to 0.50 in dense fluidized beds. The meso-scale structures, which take the form of clusters and streamers in dilute gas-solid suspensions and bubbles in dense gas-solid suspensions, coalesce and break up frequently. Both clusters and bubbles are found in turbulent fluidized beds. In these flow problems, it is impractical to resolve every bubble and/or cluster, as it requires grid sizes of the order of ten particle diameters (~1 mm), which translates to a billion nodes even for a relatively small reactor with a volume of 1 cubic meter. A more practical approach is to simulate the dynamics of only large bubbles and clusters using coarse grids and account for the effects of smaller unresolved structures through suitable sub-grid closures. Even though this may appear obvious, such sub-grid models are not available at the present time!

The current state of affairs in gas-solid flow hydrodynamics is illustrated by the following. It has now been conclusively established that clusters and streamers are formed in gas-particle flows in vertical risers. They are found more frequently near the tube walls so that, on an average, particle concentration is larger near the wall region. A consequence of such segregation is that the average velocity of particles and gas in the wall region can be downward even though the net flow is in the upward direction. Segregation and recirculation give rise to poor contacting between particles and gas, and appreciable effective axial dispersion, respectively. A mechanistic explanation of particle segregation in riser flows is still elusive, and we are not in a position to predict the effect of scale-up on the extent of segregation in riser flows even for the case of monodisperse particles.

There is reason, however, to be optimistic about our ability to model these type of flows. Tremendous progress has been made over the past two decades on the formulation of a continuum model (commonly referred to as the kinetic theory model) for a system of uniformly-sized particles in a gas, where the particles interact only through the fluid and binary collisions (Gidaspow, 1994; Koch and Sangani, 1999). Meso-scale structures in gas-solid flows illustrated on the cover of this issue, obtained in highly resolved simulations of a kinetic theory model, arise through an inertial instability associated with the relative motion between the gas and the particles and through inelastic collisions. (Work is under way in several research groups to include the effect of particle-size distribution in this model.)

One of the challenges ahead is to understand how meso-scale structures (see figure on the cover of this issue and its caption) scale with domain size, so that one can devise rational models for the effects of unresolved clusters and bubbles on the structures resolved in coarse-grid simulations. Computational experiments suggest that it should be possible to identify simple scaling rules (e.g., Figure 1 which illustrates that cluster size scales with domain size). Diffusion of species, momentum, and energy occurs predominantly through convective processes associated with these small structures. Therefore, grid-size and (macro-scale) strain-rate dependent drag coefficients, effective normal and shear stresses, effective dispersion coefficients for species and energy transport, and even reaction rate models, are essential. Yet, none of these are available at the present time even for the simple case of uniformly-sized particles. Understanding how particle-size distribution affects these meso-scale structures and figuring out how to account for them in the sub-grid models represent formidable challenges ahead of us.

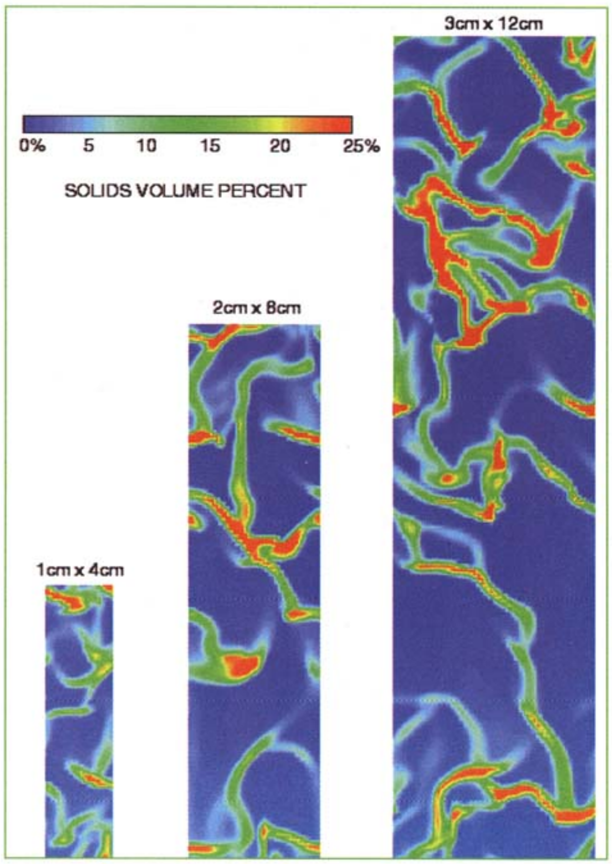

Figure 1. Instantaneous snapshots of particle volume fraction distribution obtained in 2-D simulations in periodic domains of different sizes. 75 μm particles fluidized by air. Mean particle volume fraction=0.05. Kinetic theory model. The larger the domain, the larger are the clusters and streamers.

Why is it hard to develop models for the effects of unresolved structures? Part of the difficulty stems from the fact that gas-particle suspensions with appreciable particle loading are too opaque to permit nonintrusive optical measurements. Nonintrusive measurement techniques such as capacitance, impedance and γ-ray densitometry tomographies, and radioactive particle tracking are being developed (e.g., Chen et al., 1999; George et al., 2000); presently, they do not afford adequate resolution to provide direct clues on sub-grid models. Computational experiments represent the most promising avenue, at least in the near-term, to learn more about small-scale structures in these flows.

In very dilute gas-particle suspensions, the sub-grid eddies are largely driven by macro-scale shear and modified slightly by the particles. The sub-grid scale structures in the present problem arise because of local instability and are modified by macro-scale shear. For example, sub-grid viscosity is proportional to macro-scale strain rate in single-phase turbulent flow, while in the present problem it appears to be inversely proportional to rate of strain. So, fresh thinking is needed to identify and rationalize scaling.

Gas-liquid flows in bubble columns

A state of nearly uniform bubbling may be obtained only through uniform distribution of small bubbles at very low gas superficial velocities. However, this state is stable (if at all) only in a very narrow range of conditions, and gives way to nonuniform distribution of bubbles and coherent structures at higher gas superficial velocities. In columns equipped with a small number of orifices for gas entry (which is typical), the coherent structures take the form of meandering (in 2-D, Mudde et al., 1997) or swirling (in 3-D, Pfleger et al., 1999) motion of ascending bubble plume and descending liquid-rich vortices. Coalescence of bubbles inside the rising plume produces larger bubbles that accentuate the meandering/swirling motion. These coherent structures give rise to appreciable axial mixing in the liquid-phase and lateral variation of bubble-phase fraction and gas- and liquid-phase velocities. Churn-turbulent flow, involving vigorous coalescence and breakup of bubbles, occurs at higher gas velocities, where liquid recirculation, liquid-phase axial mixing, and lateral nonuniformities are more pronounced. A bimodal distribution of bubble size (fast and slow bubbles) becomes clearly evident in this regime, and the column diameter affects the flow characteristics appreciably (Krishna et al., 1999).

Highly resolved transient simulations of two-fluid model equations with simple closure relations have revealed that the meandering and swirling flow of bubble plumes are robust flow characteristics (Pan et al., 2000; Pfleger et al., 1999). Many challenges lie ahead of us before a reliable and practical hydrodynamic model can be assembled for bubble column reactors. Experimental data on rise velocities of isolated bubbles in liquids, covering a broad spectrum of physical properties and bubble sizes, are now available, and these have been successfully correlated (Fan and Tsuchiya, 1990). On the other hand, much less is known about quantifying interphase drag in bubble swarms. Even in the case of uniformly-sized bubbles, swarms of spherical and spherical cap bubbles manifest qualitatively different dependence of drag coefficient on bubble fraction. The presence of other bubbles hinders the rise of spherical bubbles, while the opposite is the case for spherical cap bubbles. Expressions for drag force that account for this effect are just beginning to appear in the literature (Krishna et al., 1999). Validated closure models for added mass force that are applicable for swarms of spherical cap bubbles are not available. The situation is no different when we consider bubble-induced stresses.

Detailed investigation of flows of bubble swarms to extract constitutive relations for continuum two-fluid models appears to be within reach. Bunner and Tryggvason (1999) have stimulated recently 3-D flows of a swarm of nearly spherical bubbles in a periodic box through a front tracking method. Other numerical schemes such as volume of fluid method (Li et al., 1999) and lattice Boltzmann simulations are also being used to tackle this problem.

Perhaps, the greatest challenge in gas-liquid flow is the treatment of coalescence of breakup of bubbles, and the effects of these processes on the species, energy, and momentum transport processes. Simple models for maximum stable bubble size and bubble-size distribution, based on liquid-phase turbulence characteristics, have been proposed in the literature (Prince and Blanch, 1990). Detailed numerical simulations, which can be used to assess and improve these models, are sorely needed. Figure 2 shows a snapshot of a lattice Boltzmann simulation of a rising swarm of bubbles in a 2-D periodic box, subjected to oscillatory shear. Such simulations can yield computational data on rates of coalescence and breakup, mean flow and turbulence characteristics, and interfacial area, which can form the basis for improved closures for effective interfacial interaction force and stresses in the presence of active coalescence and breakup in a (micro-scale) two-fluid model.

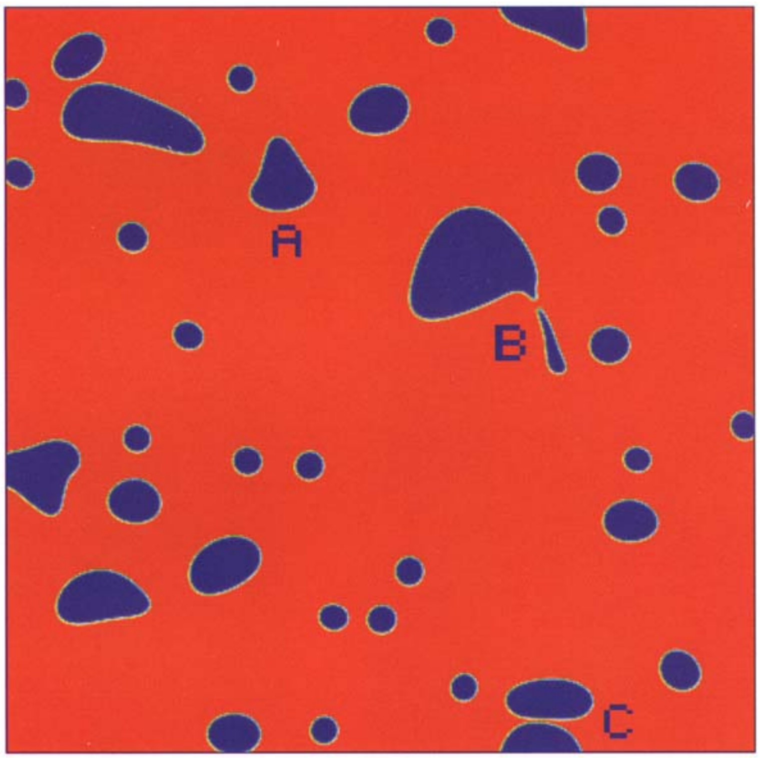

Figure 2. 2-D lattice Boltzmann simulation of rise of a bubble swarm in the presence of an oscillatory shear. Periodic domain. (A) a bubble pair that has just coalesced; (B) a bubble that has just broken up; (C) a bubble pair that is about to coalesce.

There is no doubt that such a model will manifest instabilities and lead to structures at the meso- and macro-scales, just as we discussed earlier in the context of gas-solid flows. Thus, the issue of appropriate sub-grid models for coarse-grid simulation of the continuum equations arises in bubble columns as well, but at the moment remains unexplored.

Gas-liquid-solid systems

All the issues discussed in the context of bubble columns arise in gas-liquid-solid three-phase fluidized beds, where the particles influence the bubble dynamics significantly (DeSwart et al., 1996; Li et al., 1999; Luo et al., 1997), which further complicates modeling.

Understandably, the state-of-the-art here is more primitive than in the case of gas-liquid bubble columns.

Gas-liquid flows through packed beds manifest a rich variety of spatial and temporal nonuniformities (Al-Dahhan et al., 1997). Trickling-to-pulsing transition, arising through an inertial instability, is a robust feature of continuum hydrodynamic models. Simple models of interphase interaction have been deduced from pressure drop and holdup data, but little is known about effective stresses. Detailed simulation of gas-liquid flow in pore networks, with an eye towards extracting constitutive relations for the continuum model, is an important challenge ahead of us.

To sum up

The current trend in modeling the performance of multiphase flow reactors is to integrate detailed chemistry and transport models. The main challenge is that the state of uniform flow is unstable and structures form over a wide range of length and time scales. As the macro-scale flow structures determine the large-scale mixing behavior, the ability to simulate them accurately and identify design strategies to manipulate them are especially valuable. Continuum hydrodynamic models have been formulated by averaging over details at the length scale of few particle (or bubble) diameters, but many questions still remain. While much work is in progress to address these questions, very little is being done to develop models that allow us to probe the macro-scale structures directly. Such coarse-grid models, coupled with chemistry, represent the most promising next-generation reactor models.

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