REPORT

Workshop on Scientific Issues in Multiphase Flow

to
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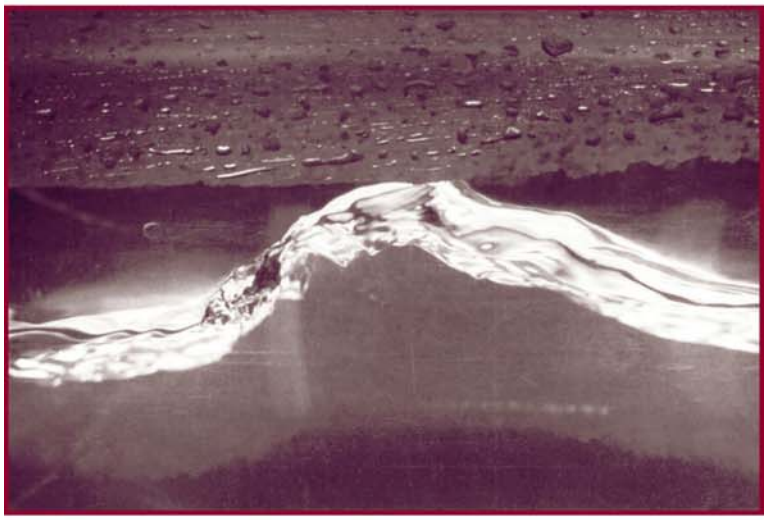

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FOREWORD

During 2001, I discussed with the United States Department of Energy the need for a Workshop on Multiphase Flow. The goal was to define basic physical issues whose resolution would improve scientific understanding. An inquiry was sent to 168 people in order to solicit ideas. A proposal was written, and accepted, which was based on the response to this inquiry and which recognized contributions made at previous workshops in Gaithersburg, 1985, Rensselaer Polytechnic Institute, 1987, and London, 1992.

Four study groups were formed to address areas of interest: (1) Phase Distribution in Gas-Liquid Flows (Theo Theofanous, chair, Thomas J. Hanratty, co-chair, Jean-Marc Delhaye, Geoffrey Hewitt, Mamoru Ishii, Daniel Joseph, Richard T. Lahey, Andrea Prosperetti, Akimi Serizawa); (2) Disperse Flow (Sankaran Sundaresan, chair, John Eaton, co-chair, Donald L. Koch, Michel Louge, Julio Ottino, Olivier Simonin, Jennifer S. Curtis, Harry L. Swinney); (3) Computational Physics (Andrea Prosperetti, chair, Gretar Tryggvason, co-chair, S. Balachandar, Shiyi Chen, Lance Collins, Howard Hu, Martin Maxey, Theo Theofanous); (4) Microphysics (Jean-Marc Delhaye, chair, John McLaughlin, co-chair, Vijay Dhir, Truc-Nam Dinh, Mostafa Ghiaasiaan, Gad Hetsroni, Juan Lasheras, Ellen Longmire, Rolf Reitz, Peter C. Wayner). A website was created so as to obtain a wide range of inputs.

Summaries of the deliberations of the Study Groups were presented at a meeting held at the University of Illinois in Champaign-Urbana on May 7-9, 2002. Short presentations were made by members of the Study Groups in support of their conclusions. The proposed research thrusts were further developed by the 47 attendees.

This report summarizes the findings of the Workshop and presents final position papers of the Study Groups. These findings were formulated through interactions amongst the chairs and co-chairs of the different Study Groups. Contributions, by individuals, to the Study Groups and to the website, which are cited in this Report, were distributed in a proceedings volume at the Workshop. Present plans are to publish these in *Multiphase Science and Technology*. The scope had to be limited because only thirty-two people were involved with the preliminary studies. Flows at low Reynolds numbers for which inertia is not important are not treated. The main emphasis is on gas-liquid and solid-fluid flows. (This choice was not meant to minimize the importance of liquid-liquid flows and of three-phase flows.) Granular flows are considered, mainly, in the context of describing concentrated fluid-solid flow systems.

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Abstract

This report outlines scientific issues whose resolution will help advance and define the field of multiphase flow. It presents the findings of four study groups and of a workshop sponsored by the Program on Engineering Physics of the Department of Energy.

The reason why multiphase flows are much more difficult to analyze than single phase flows is that the phases assume a large number of complicated configurations. Therefore, it should not be surprising that the understanding of why the phases configure in a certain way is the principal scientific issue. Research is needed which identifies the microphysics controlling the organization of the phases, which develops physical models for the resultant multi-scale interactions and which tests their validity in integrative experiments/theories that look at the behavior of a system. New experimental techniques and recently developed direct numerical simulations will play important roles in this endeavor.

In gas-liquid flows a top priority is to develop an understanding of why the liquid phase in quasi fully-developed pipe flow changes from one configuration to another. Mixing flows offer a more complicated situation in which several patterns can exist at the same time. They introduce new physical challenges. A second priority is to provide a quantitative description of the phase distribution for selected fully-developed flows and for simple mixing flows (that could include heat transfer and phase change). Microphysical problems of interest are identified – including the coupling of molecular and macroscopic behavior that can be observed in many situations and the formation/destruction of interfaces in the coalescence/breakup of drops and bubbles.

Solid-fluid flows offer a simpler system in that interfaces are not changing. However, a variety of patterns exist, that depend on the properties of the particles, their concentration and the Reynolds number characterizing the relative velocity. A top priority is the development of a physical understanding of inertial instabilities which give rise to structural features that have a large range of scales. Important microphysical problems are the understanding of particle/particle interactions, particle/boundary interactions that include the effect of wall roughness, and the influence of particles on fluid turbulence. These behaviors can differ depending on the characteristics of the particles, their size distribution and their concentration. For large concentrations, such as exist in granular flows, instabilities associated with particle-particle interactions often cause separations in systems which are not homogeneous. These instabilities are not well understood.

Appropriate averaged equations could provide a way to use an understanding of the microphysics obtained in simple systems to describe more complicated flows. The formulation of these equations presents physical challenges since the structure of the phase distribution could affect the choice of averaging methods and closure relations. Universal computational approaches appear to be out of reach at present.

1. A road map for scientific understanding of multiphase flow

A physical understanding of flows in which more than one phase is involved offers problems of far greater complexity than encountered with single-phase flows. The reasons are that the phases do not uniformly mix and that small-scale interactions between the phases can have profound effects on the macroscopic properties of the flow. Our ability to predict the behavior of these flows is a limiting factor in scaling, in analyzing heat transfer systems, in developing new technologies, in analyzing chemical reactors and in avoiding and managing accidents. Scientific advances would have an impact on interests of the Department of Energy, such as high power electronics, combustion, power systems, global warming, nuclear and chemical waste cleanup, solids handling, and transportation of petroleum products.

Considerable progress has been made in recent years in the development of instrumentation, in the use of direct numerical simulations, in the understanding of small scale physics and in the development of computational tools that have the possibility of describing the behavior of complex systems. An opportunity now exists to capitalize on this progress by addressing the problem itself: multiphase flow behavior. This realization motivated the organization of a workshop to define the basic scientific problems whose solution would have a major impact on developing the field of multiphase flows.

An outcome of the deliberations from this workshop is the development of a road map for basic research. The starting point and, perhaps, the most important scientific issue is to understand the complexity that characterizes multiphase flows. This would involve a description of how the phases organize in space and how this organization is related to the microphysics. A number of configurations can be realized for a given system, depending on operating conditions such as the flow rates and the physical properties of the phases. The tasks, then, are to predict changes of the configuration and to develop a quantitative understanding of how the phases distribute for a given configuration.

The solution of this problem involves the discovery of the small scale interactions between the phases which are controlling their distribution in space, studies of these interactions in carefully controlled laboratory experiments or direct numerical simulations, the development of physical models, and the testing of these models in integrative experiments.

Experimental studies present a number of difficulties because probes can interfere with the flow and because multiphase systems can be optically opaque. Major efforts to develop and deploy new instrumentation would be most useful. Techniques of interest are small angle scattering, X-ray and neutron tomography, nuclear magnetic resonance imagery, high-speed infrared thermometry, various microprobes, uv laser-induced fluorescence. Pattern recognition software includes the application of artificial intelligence methods such as neural networks, genetic algorithms and Bayesian-based image processing techniques. However, of equal importance is the availability of well designed and operated flow facilities that can investigate a range of equipment sizes, flow conditions and fluid properties.

One goal is the development of equations (analogous to the Navier Stokes equations for single phase flows) that can describe the organization of the phases both in simple and in complex situations. The basic equations of fluid dynamics, the equations of motion of particles or of bubbles, and the equations of interfacial dynamics are applicable. However, for most engineering systems, solving these equations will be impractical for the foreseeable future and their use will be limited to scientific studies. Therefore, one must employ equations which

describe the behavior of a system in an average sense. The development of averaged equations, which use the correct microphysics in the required closure laws, is a central problem.

The approach outlined above would seem to be obvious since it simply acknowledges that the first order problem is to understand the complexity that makes the analysis of multiphase systems so difficult. Yet, the scientific community has not given this issue the attention it deserves. For example: The flow regimes observed for gas and liquid flowing in a long pipeline were identified over 40 years ago, and empirical correlations of such quantities as the pressure drop and liquid holdup have been found to be more successful if they are tailored to a particular regime. However, validated physically based models that describe the transition from one regime to another are not available. Here, validation implies the direct verification of the postulated physical mechanisms and the testing of the predictions over a wide range of conditions.

A dramatic example of how large-scale phenomena are controlled by small-scale physics is the sensitivity of the behavior of a dense fluidized bed to the properties of the particles and to the polydispersity of the particles. Another is the change of an annular flow in a horizontal pipe to a stratified flow through the addition of very small amounts of drag-reducing polymers (Al-Sarkhi & Hanratty, 2001). The annular pattern is observed in gas-liquid flows at large gas velocities. Part of the liquid flows along the wall as a film and the rest as drops entrained in the gas. The additives destroy disturbance waves on the wall film. Because of this, atomization ceases and the ability of the wall film to climb around the circumference is diminished

2. Dilute disperse flows

2.1 *Phase configurations*

Disperse flows of solid particles, bubbles and drops are central to the analysis of multiphase flows in that they are often used as a prototype for developing general computational approaches. They are found in nature and technology (sand and rain storms, coal combustors, paint sprays, cyclone separators, liquid fuel combustors, chemical reactors, grain conveying). The goals of theoretical analyses include the prediction of the particle distribution in space, local volume fractions of the dispersed phase, the relative velocities of the phases, particle turbulence, fluid turbulence, and deposition rates. The motion of the particles is controlled or strongly affected by the relative motion of the phases. The idealized picture of uniformly dispersed spheres, which are interacting with one another, is rarely realized. For example, the solids can often distribute close to the wall in an upwardly flowing gas-solid mixture. Such distributions of the solids are a serious consideration in the operation of catalytic reactors, since the efficiency of the contacting is affected.

When the relative motion is large enough, inertial instabilities can give rise to mesoscale structures (Agrawal *et al.* 2001). This behavior is manifested by the appearance of clusters of particles or bubbles (Serizawa & Tomiyama 2002). Under some circumstances the particles can assume large scale turbulent motions that do not appear to reflect directly the turbulence patterns in the fluid.

For the cases of dispersed drops and bubbles additional complications arise because the interface between the fluid and the particles can change due to deformation, breakup and coalescence. The interfacial area density (that is, the area per unit volume) is an important

variable that captures some of these effects (Kocumustafagullari & Ishii 1995; Delhaye 2001). However, other characteristics such as drop or bubble size distribution also need to be considered.

The understanding of the spatial distribution of the particles, the formation of large scale structures and the evolution of the interfacial configuration in fluid-fluid flows become central issues. This understanding requires the identification/study of the controlling microphysics and the development of a theoretical framework to describe the observed behavior. Some progress has been made in this direction. However, in many situations (particularly, in complex flows) the organizing principles have not yet been discovered.

The above discussions have focused mainly on the effect of the fluid on the particles. However, the particles can cause increases or decreases (Kulick *et al.* 1994; Hetsroni 1989) in the fluid turbulence, even at very small concentrations. A complete understanding of these effects is not available.

2.2 *Particle tracking*

Much progress has been made on the analysis of dilute particle laden flows by studying the trajectories of a large number of particles that are introduced into a direct numerical, or large eddy, simulation of a turbulent flow. When the interaction of particles and the influence of particles on the fluid turbulence are not taken into account the results essentially represent possible trajectories of small single particles. Changes of fluid turbulence have been calculated by back momentum coupling through a point-force approximation.

Invariably these methods assume the particle Reynolds number is small, that the particle size is less than the smallest turbulent scale and that drag laws in non-turbulent fluids can be used. However, in many contexts these conditions are not satisfied and it is important to have a better understanding of fluid-particle and particle-particle interactions when this is the case. Hydrodynamic and collisional interactions of particles can become important at volume fractions as small as 10^{-4} so that one needs to be cautious when neglecting their effect. More work is warranted to use the particle tracking approach to study the basic physics and, in particular, to understand the effects of particle-particle interactions on the spatial distribution of the particles. The development of an improved understanding of the physics of particle-wall interactions (that includes the effect of wall roughness), of particle-particle interactions, and of the effect of particles on the fluid turbulence emerge as major scientific problems that need to be addressed further.

The performance of particle tracking studies in a turbulent field that is generated by a direct numerical solution of the Navier Stokes equations has the disadvantage that the size of the computations can be prohibitive if a large range of variables needs to be explored. Furthermore, the calculations are limited to small Reynolds numbers and to simple flows. An alternate approach is to use large eddy simulations to represent the fluid field. Perhaps, the simplest approach is to develop a stochastic model for the turbulence seen by the particles that takes account of the effect of the particles on the turbulence in an average sense. This approach has not been used with great confidence because direct testing of the stochastic models has not received enough attention. However, the possibility of comparing them with direct simulations or detailed experiments provides motivation for more work in this direction.

2.3 *Eulerian methods*

Ultimately, Eulerian methods need to be developed if practical calculations are to be carried out. Impressive progress has been made in recent years by using methods outlined in Section 2.2 to simulate the behavior of disperse flow. One of the more advanced calculations is a direct numerical simulation of sedimenting particles in a non-turbulent fluid in which the drag of the fluid on the solid particles is not modeled but determined from the calculated velocity field of the fluid (Hu *et al.* 2001). Comparable simulations have been done for disperse bubbly flows (Bunner & Tryggvason 2002). Not enough has been done to use numerical experiments such as these to develop the physics needed to carry out Eulerian calculations.

As indicated in Section 1, Eulerian methods, necessarily, involve the development and use of appropriately averaged momentum, mass, and mechanical energy equations. One of the more popular approaches is the "two-fluid" model which implies two interpenetrating streams. The development of these equations and the specification of closure relations continue to offer a theoretical challenge (Prosperetti 2002).

However, for fully-developed flows, a simpler approach that directly relates the distribution of the dispersed phase to the microphysics is of interest, particularly in identifying the controlling physics. For example, consider a horizontal disperse flow of drops or solids in a channel – such as observed in the core of a gas-liquid annular flow. A theory for the spatial variation of the concentration of drops could be developed by equating the diffusion velocity, $= -\varepsilon\, dc/dy$, to fluxes associated with gravitational settling, turbophoresis, forces due to particle-particle interactions and forces due to the existence of mean velocity gradients. In dilute disperse flows the deposition constant is mainly defined by the "free-flight" flux to the wall and the gravitational settling. Models are needed for these different fluxes, for the relation of the particle turbulence to the fluid turbulence, and for free-flight deposition.

3. Gas-liquid flow regimes

3.1 *A problem in complexity*

In a general multifluid system, the interfaces are free to deform, breakup, or coalesce. While topologically unconstrained, the forces in such systems cause a self-organization into spatio/temporal patterns, defined by fluid distributions and physical phenomena that are observed over a large range of length scales. Since the behavior of these systems depends on the pattern, its prediction is the overriding scientific goal. Scientific understanding will be greatly enhanced if we recognize the task as being an issue of complexity that involves the definition of the organizing principles that govern these patterns.

The starting points are studies of quasi fully-developed adiabatic flow in pipes with different inclinations and of well-defined mixing flows (that can include heat transfer). These flows are important in practice and they are simple systems in which the controlling physics can be studied. The enabling tools are dynamical numerical simulations and appropriate local and global instrumentation that can provide quantitative information about flow patterns.

In the long term it is desirable to use newly acquired physical insights to analyze more complex systems. This will, necessarily, require the development of imaginative computational approaches.

3.2 *Quasi fully-developed flows*

Some examples of fully-developed adiabatic flows are the bubbly (discussed in Section 2), annular, slug, stratified, and churn patterns.

The annular pattern is one for which part of the liquid flows along the wall and the remainder as drops in the gas. It is extremely important (particularly in heat transfer problems) but its description offers a number of physical challenges which include the development of theories for the rate of atomization, the rate of deposition and drop size. At large liquid flows a wispy annular regime occurs in vertical pipes whereby the liquid in the gas core forms agglomeration structures, which are poorly understood. In horizontal flows gravity causes the liquid in the film and the drops in the core to distribute asymmetrically. A quantitative description of these effects is extremely important, particularly in heat transfer applications where one is concerned about dryout of the wall film. Such descriptions are not available because the turbulent processes which oppose the influence of gravity are poorly understood.

The slug pattern, which exists in horizontal or near-horizontal applications, is of interest because it can be accompanied by large vibrations in equipment. It is characterized by the intermittent appearance of aerated slugs of liquid that move at velocities that are slightly larger than the gas velocity. (They, therefore, have large momenta.) There is a need to predict when this behavior will occur and the distribution of slug lengths. These, in turn, require an understanding of how slugs are formed and a model for the slug which describes the rate at which liquid is shed and the influence of slug length on stability.

A stratified pattern exists in horizontal pipelines whereby all of the liquid flows along the bottom of the pipe and the gas, cocurrently to it. It would seem to be the simplest pattern. Yet, considerable errors can be made in predicting the liquid holdup, the pressure drop and the liquid flow at which a transition to slug flow occurs. The main reason for this is that sound theories relating the drag of the gas on the liquid to the wave pattern on the interface are not available.

The idealized bubbly flow pattern, observed in vertical pipes, is described in Section 2. As the volume flow of gas increases, coalescence causes the appearance of cap-like bubbles. In relatively small diameter tubes bullet shaped (Taylor) bubbles, which fill the whole tube, are formed. At still larger flows this gives way to a churn pattern. A liquid film exists at the wall. Portions of the film are carried downward by gravity and other portions are carried upward by surface waves. A haphazard flow of the gas-liquid mixture exists over the main part of the pipe cross section. Churn flow can be different in large diameter pipes or large vessels, where Taylor bubbles are not formed and wall films are not an important consideration. Not enough attention has been given to the churn pattern, particularly when we consider that it usually exists over a wider range of flow conditions than does bubbly flow.

3.3 *Flow regime transitions*

The development of an improved physical understanding of transitions from one flow regime to another could be the most important problem that needs to be addressed. Success

would provide a basis for developing a much needed sound physical theory for predicting flow patterns. It would also establish basic physical laws that define the science of multiphase flow.

Early workers presented maps, with coordinates such as the superficial gas velocity and the superficial liquid velocity to define flow patterns. This empirical approach has failed to produce a general method to predict transitions. Taitel & Dukler (1976) were the first to attempt a general analysis in which physical mechanisms were used. This work has had a strong impact and its predictions are often used – even though they are unreliable because the physics is incorrect or only partially correct.

Some progress has been made in predicting the transition from a stratified flow to a slug flow by considering the stability of a stratified flow and the stability of a slug (Woods & Hanratty 1996). The main task for completing this analysis is to put together a synthesis of the work that has been done and to test it over a range of conditions.

Unfortunately, our understanding of the transitions amongst other flow regimes is not so well developed.

3.4 *New issues introduced in mixing flows*

Mixing flows are common in a large number of applications that involve gas-liquid systems. These include jet breakup, developing flows in short pipes, pool boiling, bubble reactors, and inertial-confinement fusion. A general approach to these types of problems is desirable. A starting point is the incorporation of an understanding of the microphysics obtained from studies of quasi fully-developed flows in pipelines into an appropriate computational scheme. However, additional problems arise that are not seen in fully-developed pipe flow. For example, more than one pattern can exist in the same field so one must be able to define local patterns from internal variables.

There is a need to carry out studies of well conceived mixing flows to test the generality of theories developed in pipe flows and to understand new problems that arise in more complex flows. A characteristic of mixing flows is that topological changes can occur over relatively small spatial regions. The understanding of the behavior of these large scale discontinuities introduces a new element of considerable importance (that needs to be understood) because the behaviors of the flow on the two sides could be qualitatively different. When the mean flow is normal (e.g., in pool boiling, shock-induced mixing, and bubble columns) the discontinuity sweeps into the mean flow and its ensemble-averaged configuration changes with time. When the flows on the two sides are parallel (e.g., annular flow and flow boiling) the ensemble-averaged behavior need not change with time.

Examples of engineering applications where large scale discontinuities play a role are the shattering of fuel drops in internal combustion engines and liquid-fuel detonation engines, jet atomizers, and the three phase flows (oil-water-natural gas) that occur in long distance hydrocarbon pipelines. In the latter the mixing of the liquid phases can lead to the formation of an emulsion of very high viscosity, which can restrict the flow. A related problem is a phase inversion where the continuous phase in the emulsion changes.

3.5 *Numerical experiments*

As mentioned in Section 3.3, recent advances in direct numerical simulation offer the opportunity to carry out experimental studies of the microphysics of gas-liquid flow, which are

not possible in the laboratory. The development of methods to track interfaces that include the physical processes of coalescence and breakup is particularly exciting. Other studies of interest are wave growth and atomization, vapor occlusion in films, some aspects of nucleate boiling, flow patterns in slugs, solidification, effect of polymer or surfactant additives, and annular flow liquid film dryout at critical heat flux (CHF).

For example, theoretical work on the rate of atomization and on the transition from a stratified flow to a slug flow are, largely, based on linear theory. There is a need to extend this work to include nonlinear effects and the possibility of bifurcations. When one considers the stability of a stratified flow in order to predict the initiation of slugging, linear theory cannot tell whether sub-critical bifurcations (i.e., multiple stable solutions) will occur. Experiments and numerical simulations are urgently needed to determine the outcome of these instabilities.

3.6 *Averaged conservation equations*

Section 3.3 has outlined the use of appropriately averaged equations to describe the velocity field in disperse flows. In gas-liquid systems these equations have been mainly developed for bubbly flows. These allow for breakup and coalescence and a change of interfacial area that does not occur in solid-fluid flows. A two-fluid approach is generally employed in which separate averaged equations are used for the gas and the liquid. Some success has been experienced in using these equations to describe the mean properties of a fully developed flow in circular and wedge-shaped conduits (Lahey & Drew 2001).

The development of universal equations which describe a large range of gas-liquid flow patterns could be an impossible task. It is not the focus of this report. Nevertheless, averaged equations will be useful in demonstrating how models for the microphysics control the phase distribution. These equations will probably need to be tailored to a particular application – so their formulation is a physical challenge.

4. Concentrated solid-fluid systems

4.1 *Concentrated 'fluid-like' solid-fluid suspensions*

When the particle concentrations become large, such as exist in fluidized beds, granular flows and some slurry flows, the influence of particle-particle interactions (mediated by continuous-phase fluid motion and direct collisions) become important or dominant. Some progress has been made in analyzing these flows by using two simplifying approaches which are based on analogies with the kinetic theory of dense gases but allow for dissipative effects of fluid viscosity and inelastic particle collisions. The theory of "rapid granular flows" neglects all influence of the continuous phase. The theory of "gas-solid suspensions" adds the effect of a low Reynolds number gas. While there are some problems in accounting for particle-particle collisions and for boundary conditions, the major issue in extending these theories is to account for the influence of the continuous phase when the particle Reynolds number is large.

This can result in stronger dissipative effects associated with enhanced dissipation of particle kinetic energy and enhanced dissipation in the fluid due to turbulence. However, the most important and intriguing problem is a consideration of the instabilities associated with the finite Reynolds number (discussed in Section 2.1). This involves a physical understanding of

these instabilities, their influence on the formulation of averaged equations, and a theoretical understanding of particle turbulence. The latter provides a challenge both to the theorist and the experimentalist.

The instabilities cover a wide range of scales involving fluctuations with a spatial scale of several particle diameters to spatial scales which are of the same size as the dimensions of the equipment. The analysis of these systems necessarily involves the use of equations which average the small scale fluctuations (analogous to large eddy simulations used in single phase turbulence). The modeling of the small scale particle turbulence emerges as a major problem, particularly because instabilities make experiments on the rheology of high Reynolds number suspensions difficult.

In some circumstances, instability leads to the appearance of pockets in the continuous phase, such as observed in dense fluid beds. Then, it is probably advantageous to use two-field equations, which analyze the pockets and fluid-solid suspension separately, and appropriate conditions in the regions separating the two fields.

4.2 *Dense 'solid-like' systems*

It is important to understand the conditions leading to transition from a fluid-like to a solid-like behavior, where the particles interact through enduring contacts between each other and the boundaries. These transitions are important in granular flows and have been observed in fluidized beds. Primary problems are to understand cohesive interparticle forces, frictional contact between particles, and the stability of the system. Quantitative predictions of the performance of many devices, such as fluidized beds, spouted beds, standpipes, dense phase pneumatic conveying and mixing equipment depend on our knowledge of frictional stresses.

An interesting aspect of this problem is that interparticle forces depend on the shape of the particles, the size distribution and frictional contact. A knowledge of this dependency opens the possibility of manipulating the flow behavior by tailoring particle characteristics. Constitutive models developed in soil mechanics are a good starting point but there is growing evidence that these should be modified to bring in the effect of strain rate fluctuations on the transition between quasi-static and rapid flow regimes.

An important aspect of all these systems is the tendency of granular materials to segregate because of small differences in size, density, surface roughness and shape (Ottino & Khakhar 2000). This is a complex and imperfectly understood phenomenon of practical importance. To describe segregation we not only need models for the flow but also constitutive models for segregation fluxes under various flow states. An understanding of this issue is relevant to a wide cross section of industries.

5. Microphysics

5.1 *Identification of important microphysics*

The relevance of microphysical studies needs to be measured by improvements in our ability to describe large scale phenomena of interest. Many aspects of the microphysics have been discussed in Sections 2-4. This section gives examples of problems that have not been directly mentioned.

The coupling between molecular and macroscopic phenomena is a feature of many multiphase flows. For example, it is well known that relatively small concentrations of surfactants can promote the breakup of bubbles or drops in turbulent flows. However, there is also experimental evidence that some surfactants have the opposite effect. This coupling is a theme in much of what is presented in this section. This includes nucleate boiling, contact lines, the effects of surface active agents and drag-reducing polymers, microscopic conditions at the point of breakup or coalescence of interfaces.

5.2 *Coalescence and breakup*

An improved basic understanding of breakup and coalescence of interfaces would have an impact on a number of problems involving fluid-fluid flows. These include the prediction of bubble size and interfacial area per unit volume, drop size distribution, the breakup of liquid jets, the formation of sprays, and the atomization of flowing liquid layers. Direct numerical simulations and innovative experiments can provide information needed to formulate equations that define the rate processes.

An important consideration in developing predictive models is the microscopic conditions at points of breakup and coalescence. Breakup of drops and bubbles involves intermolecular forces acting at micro- or nanoscales. For example, if one considers a highly deformed bubble in a liquid, breakup may occur if a sufficiently thin neck forms between two portions of the bubble. Intermolecular forces between the liquid molecules are an important factor – determining when the neck pinches off and two child bubbles result. The modelling of final aspects of breakup and coalescence is particularly important in computational experiments. However, it is quite possible that instabilities dictate the initiation of breakup and coalescence so that analyses need not go down to the molecular level.

5.3 *Altering the behavior of a multiphase system*

An area of research that has not been fully exploited is the possibility of altering the behavior of multiphase systems by modifying the microphysics. Examples are the use of structured surfaces in heat transfer, the addition of fines in fluidized beds, the addition of surfactants and drag-reducing polymers. Such studies test our current physical understanding of multiphase flows and suggest technological opportunities.

The influence of surfactants on bubbly flows is an example of how molecular scale processes affect the behavior of multiphase systems (Zhang *et al.* 2001). They affect the final stages of coalescence and breakup and, therefore, bubble and drop size distributions. They also affect small wavelength waves which are important in understanding interfacial instabilities. Drag-reducing polymers affect both interfacial properties and the bulk flow. Their role in changing annular gas-liquid flows, in modifying the liquid flow and interfacial stresses in stratified gas-liquid flows and increasing the stability of slugs has been discussed in other sections of this report. A theoretical explanation of these results is needed.

5.4 *Contact lines*

An understanding of the motion of contact lines plays an important role in many aspects of multiphase flow. These include the wetting of the pipe wall in a horizontal gas liquid flow,

the stability of dropwise condensation, the behavior of gas cavities and critical heat flux in boiling, and the behavior of liquid films. The motion of contact lines depends on the local environment in ways that are not fully understood. Experiments on rapid condensation of steam onto surfaces with wettability gradients indicate that the motion of drops can be dramatically different from their motion in the absence of condensation (Daniel *et al.* 2001). Also, it has been speculated that contact line pinning occurs when a moving contact line encounters a geometrical irregularity or a hydrophobic patch (de Gennes 1985), but this has not been verified.

5.5 *Nucleate boiling*

In many situations the most difficult part of analyzing a heat transfer problem is the prediction of how the phases distribute, so adiabatic experiments can provide a basis for analysis. An example is burnout caused by the disappearance of the wall film in an annular flow. This is not the case when phase changes alter the boundary condition at the wall. Considerable work has been done on nucleate boiling and on dropwise condensation but major problems need to be resolved involving the prediction of nucleation sites, and of the effects of apparent contact angle and contact line motion on heat flux.

It is well established that nucleate boiling occurs at cavities that contain entrapped gas and that the rate of heat transfer depends on the number of active cavities. Progress has been made in understanding this process but more needs to be done, particularly for situations in which the liquid is flowing. Recent experiments (Theofanous *et al.* 2002) have shown that nucleate boiling can occur on nanoscopically smooth surfaces. Here, molecular scale surface inhomogeneities may be expected to play an important role. Clearly, the relation of nucleation and resistance to burnout to the surface characteristics needs much deeper consideration. Also studies aimed at understanding the role of surfactants in enhancing nucleate boiling (Hetsroni *et al.* 2001) could be rewarding.

5.6 *Heat transfer in microchannels*

Networks of microchannels with hydraulic diameters in the 100 microns to 1 mm range have the capability for sustaining very large transport rates in small volumes. Consequently they are becoming increasingly important. They are characterized by large effects of surface tension, of surface wetting and of contact line movement. Inertia cannot be ignored. Surprisingly, many aspects of the flow patterns are similar to those observed in macroflows. However, quantitative descriptions of the behavior are not available. For example, it would be of interest to extend our understanding of annular flow in macroflows to microflows – and, in particular, to predict liquid film dryout (i.e., CHF). Because the small channel size could alter bubble growth, it would be expected that flow boiling could be quite different from what is observed in macroflows.

6. Closure

The central issue in multiphase flow is the understanding of the mechanisms and processes which determine the spatio-temporal distribution of the phases. Successes in meeting this challenge would have an immense impact on engineering practice and, indeed, the world's economy.

As emphasized in this report, reaching this understanding requires an integration of progress made on sub-problems, such as the behavior of particles in a turbulent field, bubble dynamics, interfacial phenomena, phase change, breakup and formation of new interfaces, boundary effects, granular flows, contact line motion, nucleation, and many others. Progress can be expected to be much swifter than in the past, given the availability of better experimental techniques (e.g., X-ray tomography, infrared photography, atomic-force microscopy), more powerful computational capabilities (e.g., parallel computers, larger memory, modern algorithms), and a deeper theoretical understanding (e.g., multiscale integration, nonlinear analysis).

A feeling, broadly shared by the Workshop participants, was that the best way to proceed is to promote an understanding of the basic physics of multiphase flows. In other words, the emphasis of research in this area should change from a strictly engineering viewpoint (which has had limited success in developing general approaches) to a science-oriented one. Coupled with the remarkably powerful new experimental and computational tools that are now available, this shift of emphasis promises major advances in a relatively short time.

Progress would be further accelerated if a focused research effort could be promoted. The contemporary research enterprise is inherently complex and expensive. Encouraging researchers from different institutions to pool resources and expertise would ultimately benefit the entire community. The fostering of medium to large-scale collaborative efforts, that includes joint mentoring of students, would greatly benefit the discipline of Multiphase Flow.

Acknowledgements

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WORKSHOP ON SCIENTIFIC ISSUES
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APPENDIX 1

REPORT OF STUDY GROUP ON FLOW REGIMES IN MULTIFLUID FLOW

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APPENDIX 1

REPORT OF STUDY GROUP ON FLOW REGIMES IN MULTIFLUID FLOW

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1. Introduction

In a multifluid system, the interfaces are free to deform, break up, or coalesce. While topologically unconstrained, we find that under the influence of the relevant forces such systems tend to self-organize into spatial/temporal patterns, defined by fluid distributions and length scales. Since much of the behavior depends on these patterns, their prediction is the overriding scientific goal in pursuit of our subject. Unfortunately it is a goal that remains largely unfulfilled, despite intense investigations beginning over half a century ago.

There is an opportunity that, if taken, will prevent a similar kind of statement being made 50 years from now. The opportunity is created by tremendous advances made in infrastructure crucial to the task – instrumentation from nano and micro technologies, materials science, digital technologies, numerical simulations capability (that result from leveraging clever schemes with hardware speed and capacity). However, the opportunity will escape us unless we see the task for what it is – an issue of complexity – and unless we take steps to approach it accordingly. This would involve searching, discovering, and verifying conclusively the "laws" (or organizing principles) that govern these patterns.

It is suggested that this challenging task will require a highly collaborative atmosphere that has to be anchored in experimentation. Our interpretation of experimental results should involve theory and simulations of all kinds, from molecular dynamics, to DNS, to effective field models, and ultimately to true multiscale treatments. The aim should be understanding the key

physics behind the phenomena being studied. The necessary discoveries cannot be *a priori* guaranteed. Thus we need to allow patience with others' efforts and to have an attitude that is more collaborative and less exclusive. This may also require some structural changes by the sponsors (of research) to catalyze and promote synergisms and collaborations.

With this perspective, the individual contributions of the Task Group must be considered only as indicative; that is, we want to emphasize the goal and the process (the way of doing), rather than the specifics. The contributions are highly complementary. Hanratty (2002), Joseph (2002) and Prosperetti (2002) address mechanisms and criteria for regime transitions in fully-developed, steady-state, adiabatic pipe flows. Serizawa & Tomiyama (2002) and Lahey & Drew (2002) address the detailed characteristics of bubbly flows. Theofanous & Dinh (2002) bring out the importance of mixing flows and suggest that these, as well as disperse flows, could be viewed as the canonical components in a multiscale treatment that is focused to understand self-organization in multifluid systems. Ishii (2002) discusses interfacial area transport within a multifield formulation and Lahey & Drew (2002) consider a constitutive treatment of forces as a way of distributing the phases within a multifield model. These two approaches are complementary since the basic source terms for the formation of interfacial area density are also the source terms for the field-to-field exchanges which may, in turn, lead to flow regime transitions. Significantly, these sources must be developed from an understanding of microphysical phenomena. The above summary could suggest that there is no uniformity of opinion; actually there are considerable synergisms and complementarities, as discussed below.

2. An overall view

The overall organization indicated in Fig. 1 emerged. Each of the three rectangular boxes indicates the scope of a particular (broad) domain of investigation. Again, the aim within each of these domains is to determine the physics (laws) that govern self-organization and, by implication, transitions. At the center are the enabling tools, that is, instrumentation and dynamic numerical simulations software. The instrumentation is critical to reveal patterns and the numerical simulations are essential in interpreting what is revealed by the experiments. They are centrally positioned because what is learned in one context is available for use in another. The double arrows suggest the important interplay between specific analytic activities (scaling laws, theory, simple ad hoc models) and the numerical simulation task. As mentioned above, these central tasks include any kind of simulation tool that exists or could be developed and fruitfully applied.

The numbers indicate the order of priority. On one hand, steady, fully-developed adiabatic pipe flows are the cornerstone whose understanding is overdue. Besides being very important for practice in their own right, they also provide the simplest possible anchor for developing and testing numerical simulation tools. Moreover, we can expect that what is learned (by such simulations) could substantially aid the discovery of the organizing principles being sought. Simple mixing flows are the other important cornerstone to develop. Besides being practically important, these need to be understood before anything more complex, such as unsteady, developing flows in complex geometries can be addressed in a sound fundamental way. Since the prediction of such complex flows (Box 3 in Fig. 1) requires a panoply of dynamic simulation tools, it is suggested that developments and applications in all three areas are mutually beneficial and are indispensable features of an overall “integrative” approach.

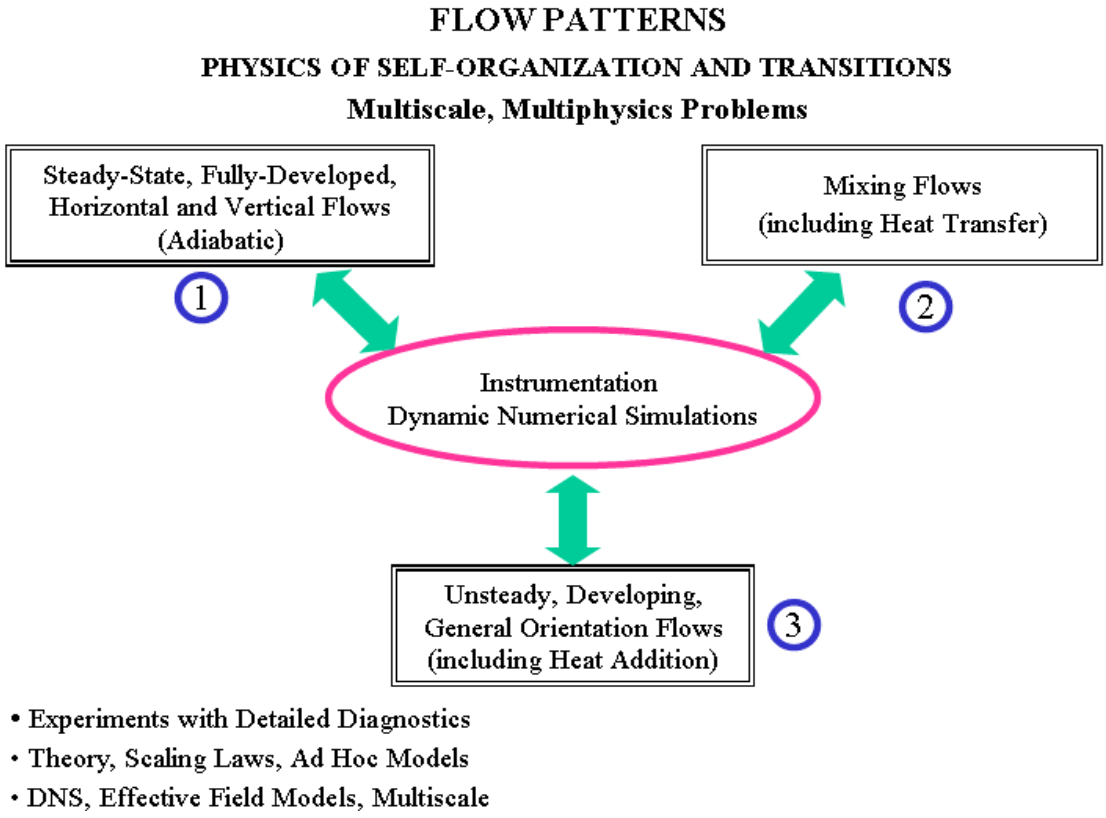

Fig. 1. Key Elements of an Overall Strategy.

The remaining three sections of this report discuss in slightly greater detail what is contained in the two basic thrusts (the two top Boxes in Fig. 1) and the enabling central thrust. The presentation is for the most general case of gas-liquid flows that can include phase changes due to heat transfer (boiling, condensation). However, the analogous and important case of liquid-liquid systems should also be kept in mind. Detailed discussions are found in contributions by different members of this Study Group that are identified in the last paragraph of the Introduction.

3. Steady, fully-developed, adiabatic flows

An overall view of steady, fully-developed adiabatic flows is depicted in Fig. 2. The top line gives a classification that is simplified by first considering dispersed and separated flows. The latter can be either (gravity) stratified or annular. Clearly there are in-between patterns. The most important of these are intermittent flows that exhibit temporally varying large scale structures. Examples are slugs (in horizontal/near horizontal or in small diameter vertical pipes) and gas/vapor-dispersed subregions (churning) in large diameter vertical flows. A key aspect of the topology is the extent of the continuity of the gas/vapor flow path, which defines the degree

of coupling between the gas/vapor (which normally has a much higher volumetric throughput) and the liquid streams. The two extremes, in this viewpoint, are dispersed and separated; the intermittent is transitional. By dispersed, we mean both bubbly and droplet flows, although it should be noted that the vapor core of annular flows normally contains drops.

STEADY-STATE, FULLY-DEVELOPED, ADIABATIC FLOWS

	Dispersed	Stratified	Annular	Intermittent
Horizontal	D	B	A	B
Vertical	C, D		A	C
Inclined	D	B	A	B

A. Phase distribution. Drop entrainment and deposition? Film shear/waves and redistribution mechanisms? Drop dynamics and gas turbulence?

B. Slug Formation and Dynamics. Interfacial shear and holdup? Mechanism of formation and sustenance? Linear or Non-Linear behavior? Role of viscosity? Surfactants and drag-reducing polymers?

C. Transition from Dispersed to Intermittent. Gradual coalescence. Limits of slug flow with channel diameter? Using dynamic response for identification of mechanisms?

D. Phase Distribution in Bubbly Flow. Coping with many types interactions generically?

Fig. 2. An overview of Element 1 in Fig. 1 and related issues.

3.1. *Phase distribution in annular flow*

Annular flows are especially important in power equipment, where a first order problem is the prediction of the fraction of liquid that flows as a film along the wall. In horizontal flows, we also need to be concerned about gravity-induced asymmetries in the local film thickness and flow rate. Dryout of this film defines a point of markedly lower heat transfer coefficients and, possibly, burnout. As Hanratty's (2002) summary indicates, we have a reasonably good qualitative understanding of the phenomena, but the quantitative treatment remains empirical. Furthermore, the data base is insufficient to claim a good understanding of the scaling laws. The treatment of annular flows mainly considers interfacial shear, and rates of entrainment of drops and deposition (which in steady, fully developed flow are the same). We don't know how entrainment occurs under varying flow conditions and fluid properties. We don't know the origin and extent of validity of the concept of a minimum film thickness, below which entrainment ceases. We don’t know the effect of a wall heat flux on the entrainment and deposition processes. We don’t understand the wispy annular flow for which agglomerative structures develop in the entrained droplet field at high liquid flows. Finally, we don't know how annular flow arises from stratified or intermittent flows, and we certainly don't have a numerical simulation that is capable of predicting this transition.

3.2. *Slug formation and dynamics*

The starting point for a consideration of slug formation is usually a stratified flow. A key need is the prediction of interfacial friction since it controls the liquid holdup and, therefore, the base flow for slug development. While some success has been realized by using descriptive approaches, a first-principles understanding of how waves grow to become slugs and how slugs sustain themselves is still lacking. Initial efforts that use linear stability theory need to be complemented by a consideration of non-linear effects in the framework of bifurcation theory (Joseph 2002), and by special purpose numerical simulations (Theofanous & Dinh 2002). The interesting effects of liquid viscosity, gas density and surface tension could help discriminate among approaches.

3.3. *Transition from disperse to intermittent flow*

As gas velocity in a vertical disperse pattern increases the two-phase flow needs to "expand", so as to provide a degree of gas-continuity in the flow path that is needed for "venting". In small pipes this gives rise to rapidly-moving slugs of liquid. In large diameter pipes, or vessels, we have something akin to channeling which, being unsteady, gives the appearance of churning. The idea that this occurs by gradual coalescence of bubbles is addressed by Ishii (2002) within the context of his interfacial area transport formulation. Prosperetti (2002) suggests other kinds of dynamic mechanisms and special experiments, with dynamically varying flows at the inlet, as a way of discriminating. Actually, before any real progress can be made, we need to carefully characterize the patterns by measured properties of the flow. In view of the need to use large scale equipment, the complexity of the patterns, and the inherently opaque setting, this is a major challenge.

3.4. *Phase distributions in bubbly flow*

Bubbly flow is the most extensively studied regime. Two different perspectives emerge at this time, and it is very important that they be reconciled. On one hand, Lahey & Drew (2002) present a fully-defined multifluid, multiphase CFD model for fully developed monodisperse flows that is shown to predict all key internal ingredients of bubbly flows, including complex geometries, such as wedge-shaped channels. On the other hand, Serizawa & Tomiyama (2002) present a comprehensive experimental study that makes evident the variability and sensitivity of such flows to entry conditions. They conclude that a proper description would require a consideration of many types of interactions, as well as a careful specification of how the bubbles are formed at the inlet. Lahey suggests that these types of interactions may be accommodated within the structure of a multifield, multifluid model. However this remains to be demonstrated.

4. Mixing flows

Mixing flows are the opposite of fully-developed flows, in that one is concerned with the very early stages of creating new multifluid/dispersed topologies. As indicated in Fig. 3, a convenient classification is in terms of the direction of mixing relative to the directions of the

mean flows involved in the mixing process. The topological changes of interest occur over relatively sharp spatial regions. Theofanous & Dinh (2002) refer to them as Large Scale Discontinuities (LSD's). When the mean flows are normal to LSD's, these surfaces (that demarcate the mixing fronts) sweep into the parent flows. When the mean flows are parallel, the LSD's may be steady, as in annular flow, or may sweep into the parent flow, as in flow boiling. Furthermore, the LSD's may be in effect stationary, as in pool or flow boiling, or they may be free to move, as in slug flow or mixing jets (both in parallel as well as in cross flow).

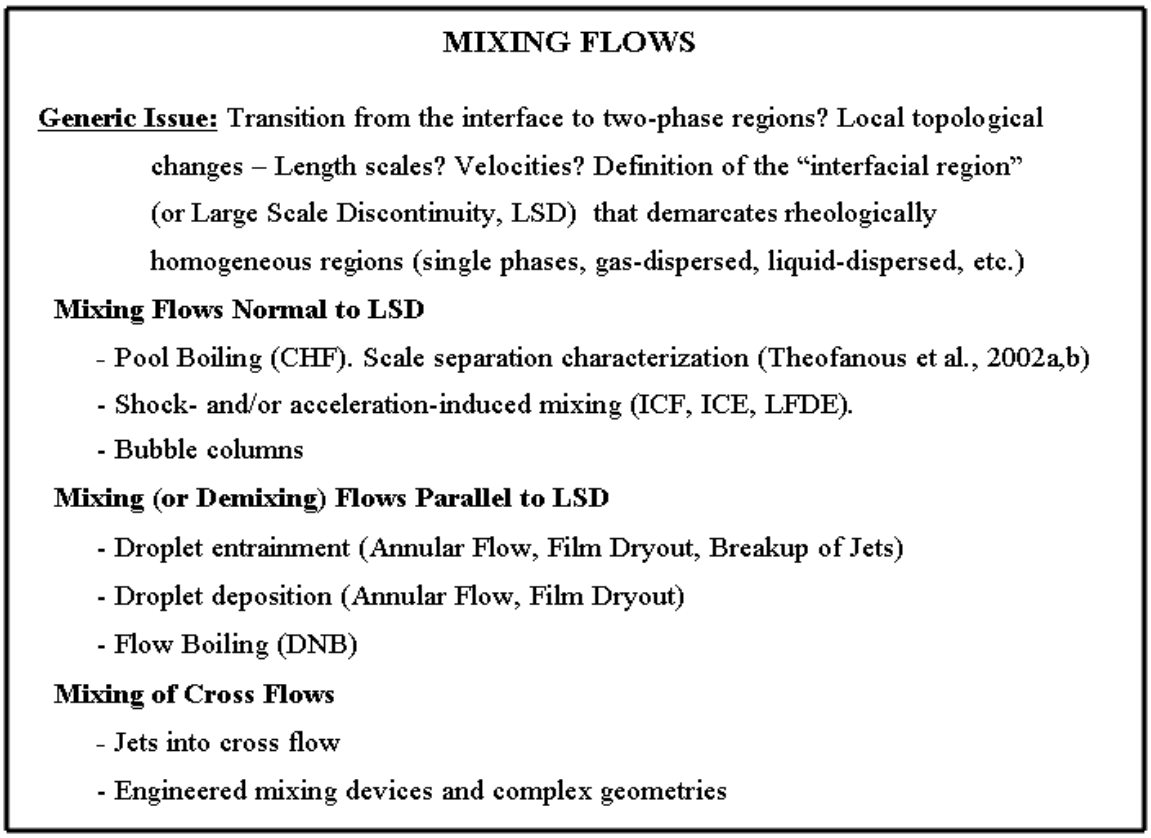

Fig. 3. An overview of the issues for Element 2 in Fig. 1.

Although not previously seen as such, classical examples of mixing flows are pool and flow boiling, and annular flow. The creations of topological changes along the LSDs constitute scientific problems of great importance for the basic understanding of these flows. Shock-induced mixing flows are a relatively new topic of interest. They occur in Inertial Confinement Fusion (ICF), Internal Combustion Engines (ICE), and Liquid-Fuel Detonation Engines (LFDE). The latter two are concerned with the shattering of fuel drops, and their mixing and reacting with a gaseous oxidant. Among many examples of an engineered mixing device an example is the jet atomizer discussed by Reitz (2002) in his contribution to the Workshop's Microphysics Thrust. Another important case, where mixing phenomena are crucial, is in three-phase liquid-liquid-gas flows. These occur in hydrocarbon recovery where oil-water-natural gas mixtures flow for long distances in pipelines. Mixing of the liquid phases can form emulsions with very high viscosity,

that restrict the flow through a pipe. A related problem is phase inversion where there is a changeover of the continuous phase.

The central issues in all of these flows are the definitions of the rate and length scale of mixing, the evolution of topological changes, and descriptions of transitions from the LSDs to the main flow regions that would be amenable to an effective field treatment. Again, while many tools are needed to understand and describe the flow patterns, special attention needs to be paid to interfacing such LSD-related behaviors to appropriate effective field computational frameworks.

5. Simulations: approaches and issues

Our view of the role of simulations in the overall task of discovering the physics of self-organization in multifluid flows has been described above. Here, we would like to emphasize the following

(a) The one-suit-fits-all idea, as embodied in the two-fluid model, is not compatible with the level of complexity of the task.

(b) It is not *a priori* clear how to do simulations that are consistent with the physics in a reasonably general way, and at a scale that is relevant practically.

As a result, we suggest that the development of appropriate simulations is a scientific undertaking of major significance that is on par with experiments. This view is consistent with the conclusions of the Thrust Group on Computational Physics, who discuss the data mining necessary to capitalize on DNS for developing constitutive laws and gaining ideas about the (small scale) organization of the phases. Further, both the Computational Physics and the Disperse Flow thrusts address, in considerable depth, issues that arise in the Effective Field (EF) framework for disperse flow simulations. Here, we only need to stress the need to simulate other (than disperse) regimes.

One approach is to capture patterns and transitions via a local application of constitutive laws within an EF (two-fluid model) framework (Lahey & Drew 2002; Ishii 2002). Ishii (2002) developed constitutive equations for coalescence and breakup events and used them to calculate interfacial area transport. Lahey & Drew (2002) propose the ensemble averaging of flow-regime-specific DNS results to help describe forces within the multi-field, multifluid model interactions that avoid the need for a separate interfacial area transport equation. Another approach is to track Large Scale Discontinuities by matching the flows on either side in a general 3D framework (Theofanous & Dinh 2002). This approach builds on the Disperse Flow formalisms and requires constitutive laws on the LSDs.

The stratified-to-slug transition in horizontal flows would make an ideal case to test these ideas and would contribute to the experimental and analytical efforts described in Section 3.

6. Concluding remarks

We believe it is important to emphasize again the role of experiments, pattern-revealing diagnostics, and pattern recognition software. The hardware includes instrumentation such as X-ray and Neutron Tomography, Nuclear Magnetic Resonance Imaging, High-Speed Infrared Thermometry, UV (laser)-Induced Fluorescence. Pattern recognition software includes artificial

intelligence methods such as neural networks, genetic algorithms, and Bayesian-based image processing techniques. However, without proper flow facilities, these techniques cannot be utilized. The availability of such facilities must be considered an integral part of the research needs in this area.

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WORKSHOP ON SCIENTIFIC ISSUES
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APPENDIX 2

REPORT OF STUDY GROUP ON DISPERSE FLOW

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APPENDIX 2

REPORT OF STUDY GROUP ON DISPERSE FLOWS

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1. Introduction

This section addresses disperse flows where the loading of the disperse phase ranges from dilute to dense. The systems considered include fluid-particle mixtures (where we consider the entire range of particle volume fractions from dilute to random close packing condition), dilute droplet laden gas flows (where the liquid droplets can essentially be treated as particles) and bubbly suspensions.

Important challenges ahead of us in the area of disperse flow include:

a) Understanding and quantitative modeling of gas-particle interactions when the particle size is larger than the Kolmogorov scale,
b) Development of averaged equations of motion and associated closure for situations where the continuous phase inertia associated with the relative motion between the continuous and dispersed phases is important,
c) Understanding the statistical characteristics of fluctuations resulting from instabilities associated with the inertia of relative motion between the phases and development of coarsened equations of motion,
d) Development of quantitative models for contact stresses in dense granular assemblies resulting from frictional and cohesive interactions between the particles, and
e) A thorough understanding of the competition between mixing and segregation in granular systems, including techniques to control these through manipulation of inter-particle cohesive interactions.

These are discussed in a greater detail below. This report groups the systems in terms of disperse phase loading levels as follows:

a) Dilute particle-laden flows, where the influence of particles on fluid turbulence and the effect of fluid turbulence on particle motion are principal issues,
b) Suspensions at modest to high particle loading levels, where the issues range from formulation of averaged (two-fluid) equations of motion and associated constitutive relations to instabilities and their consequences to suspension flow,
c) Mixing, segregation and flow of dense particulate systems with enduring contact between particles and between particles and bounding surfaces, where the particle interactions, cohesion and size/shape/density distribution influence the way particles pack and respond to imposed stresses, and segregate/mix.

2. Dilute particle laden flows

Here we focus exclusively on dilute particle or droplet-laden gas flows, where the volume fraction of particles is very small, but the mass loading ratio may be as large as unity or even greater. Strong two-way coupling, where the motion of the fluid has a significant effect on the particle motion and vice versa, is present. Interparticle collisions may also be significant.

Such dilute particle-laden gas flows are found in nature and technology, e.g. sand and rain storms, coal combustors, paint sprayers, cyclone separators, and liquid-fuel combustors. The regime may extend to fast-fluidized beds, grain conveying systems, and snow avalanches. Most or all of these flows are turbulent. While, geometric parameters and the overall Reynolds number usually are sufficient to describe the baseline single phase flow, the addition of the particle phase adds several more parameters including the mass loading ratio, the Stokes number (particle time constant/fluid time scale), the particle Reynolds number, and the particle diameter normalized by an appropriate length scale (commonly, the Kolmogorov length scale).

2.1 *Large-eddy simulations*

A large fraction of recent CFD development work for particle-laden flow is centered on extending existing single-phase large-eddy simulation or direct numerical simulation codes by the addition of particle tracking and back momentum coupling (through point-force approximation). These codes can track several million individual particles simultaneously and have recently been applied to the computation of complex geometry systems (e.g., stirred reactors) (Wu *et al.* 2001) and also in simple geometries to explore basic phenomena such as fuel droplet clustering in spray combustors (Oefelein 1998).

These codes assume that the particle Reynolds numbers are small and that the particles are much smaller than the Kolmogorov scale. However, in many applications, particle Reynolds numbers are in the range of 0.1 to 100 and the particle diameter is comparable to or even greater than the Kolmogorov length scale. An important challenge is to understand the fluid/particle interactions in such a parameter range and develop validated models (for use in the CFD codes). Specific examples of the shortcomings of the current CFD codes are illustrated below.

- Particle-tracking in these codes is done using highly simplified equations of motion; generally, the resolved fluid velocity field is interpolated to the position of the particle and an analytical or empirical drag law is applied assuming that the particle is moving in a locally uniform velocity field. This neglects the fact that the flow is turbulent and may have motions with the same scale as the particle. Published large eddy simulations, rarely, if ever, include any effect of the sub-grid scale turbulence on either the overall drag or on motions transverse to the inferred relative velocity.
- The drag laws used in most simulations accounting for two-way coupling are based on the undisturbed relative velocity. However, when two-way coupling is implemented in the code, the undisturbed velocity isn't available. Simple calculations suggest that the standard technique can cause very large particle tracking errors (Burton *et al.* 2001). New analytical research is needed to develop an appropriate way to use the self-disturbed velocity field to calculate the particle drag.
- The existing codes ignore the effect of particles on small-scale turbulence that is modeled in large-eddy simulations. However, it is difficult to argue that this effect is insignificant in flows where the turbulence is modified strongly by the particles.
- Large-eddy simulations require very fine grids near solid boundaries, and in many cases, the grid spacing is smaller than the particle diameter. Implementation of standard particle-tracking and back-coupling algorithms is clearly not appropriate.

2.2 *Fluid-particle interactions*

The issue of accurate particle drag laws which are applicable even in flows where the particle diameter is of the same order as the Kolmogorov length scale is of fundamental interest and goes beyond use in CFD codes. There is little information in the analytical, computational, or experimental literature to guide us in formulation of these drag laws. One can imagine that turbulent eddies on the same scale as the particle would have a significant effect on the drag applied on the particle and on motions transverse to the average relative velocity. The drag on a particle with Reynolds number of order ten is likely to be sensitive to the details of the particle wake. Those details could be modified significantly by small-scale turbulence. Highly resolved computations and experiments are required to assess the importance of this issue and to guide the development of models.

2.3 *Particle-particle interactions*

Collisions between particles may be significant in this flow regime. Several workers have used detailed simulations to study the statistics of particle collisions in simple turbulent flows (e.g., see Sundaresan & Collins 1997; Reade & Collins 2000), where the particles are small compared to the Kolmogorov scale. Collisional and hydrodynamic interactions between particles, when the particle size is not small compared to the Kolmogorov scale, represent fertile areas of research.

Due to the potential importance of collisions and the growing importance of codes tracking millions of particles, research into efficient models to represent the effects of collisions is imperative. Detailed experiments should be proposed to examine collision statistics in a dilute

turbulent gas flow. Three-dimensional particle-tracking velocimetry seems to offer an appropriate vehicle to study this.

2.4 *Turbulence modification by particles*

One of the most important problems in dilute particle-laden flows is turbulence modification. An early review of this phenomenon, given by Hetsroni (1989), shows the tendency for the fluid turbulence to increase in the presence of large particles and to decrease in the presence of small particles. In some simple turbulent flows, reductions of the turbulent kinetic energy by greater than 80% have been observed at mass loading ratios below 0.5 (Kulick *et al.* 1994), as shown in Figs. 1 and 2. Such dramatic changes in the turbulent kinetic energy (shown in Fig. 1) may be expected to have pronounced impact on heat and mass transfer characteristics. (For example, see Hetsroni *et al.* 2002.) Simultaneous measurements of turbulence and heat transfer characteristics of particle-laden flows should be made to explore and understand this coupling.

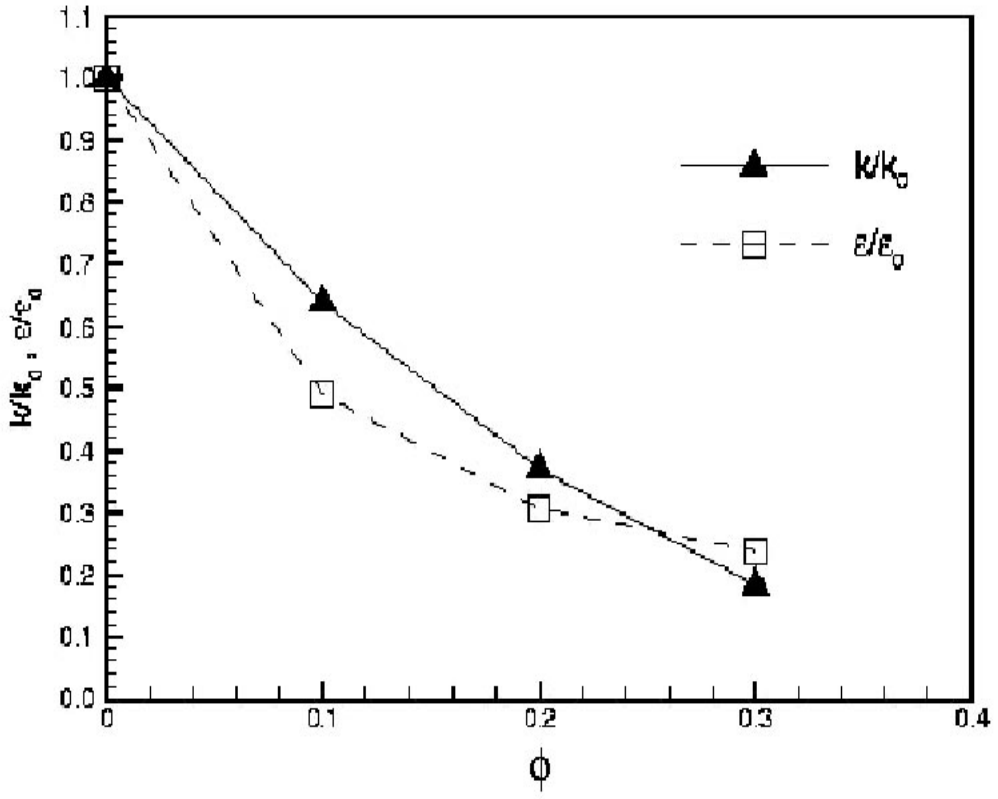

Fig. 1. Turbulent kinetic energy and rate of viscous dissipation as functions of mass loading ratio. These quantities are scaled with respect to corresponding quantities of zero mass loading. Data were obtained in the center-plane of a fully developed channel flow of air with uniform loading of 150 micron glass spheres, with resolution down to the Kolmogorov scale (but not to the particle scale). The channel flow Reynolds number based on the half width and the mean velocity was 13,800. Constant mass flow of gas was maintained. Source: (Paris *et al.*, 2001).

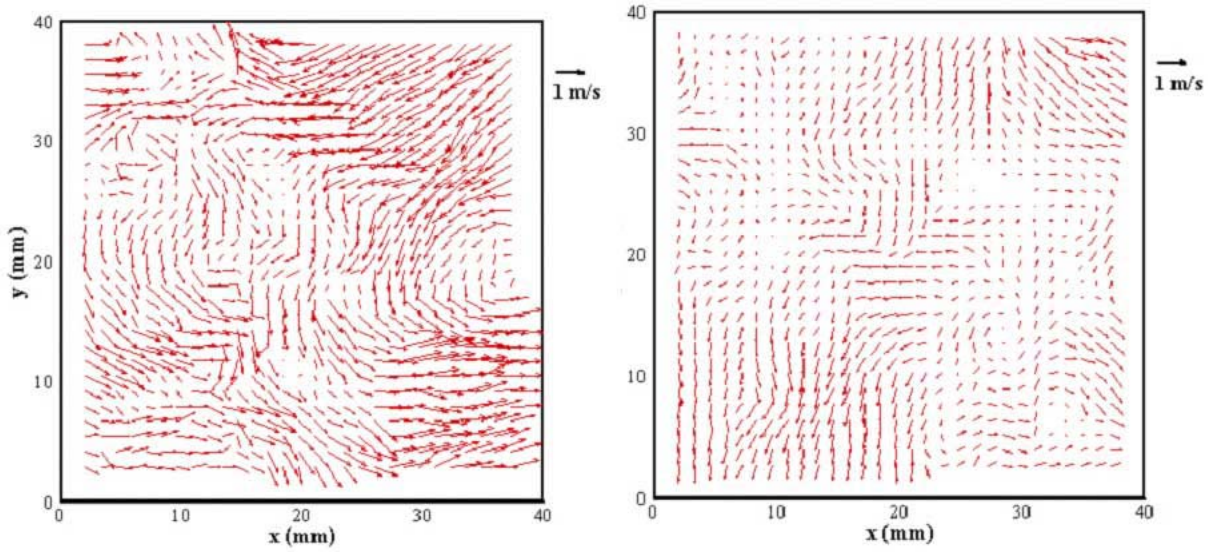

Fig. 2: Instantaneous snapshots of gas velocity fields. (a) unladen flow with a Taylor microscale Reynolds number is 470. (b) with 150 micron glass spheres at a mass loading of 0.3. Data obtained in a forced homogeneous/isotropic turbulence chamber operated under micro-gravity conditions. The scales in both figures are the same, and the strong attenuation due to the particles is evident. Particle phase velocities are not shown. Source: Unpublished work of Hwang, W. & Eaton, J.K. (2002).

Other quite similar flows reveal almost no turbulence modification up to fairly high mass loading ratios (Fessler & Eaton 1999). Some other flows containing large particles exhibit substantial enhancement of turbulence (Gore & Crow 1989; Parthasarathy & Faith 1990). Turbulence attenuation can have enormous effects on device performance, yet there is no theory or model that allows consistently accurate predictions of turbulence modification. The physical mechanisms are so poorly understood that experts in the field cannot predict if turbulence attenuation will occur in a given flow.

Turbulence modification is apparently a strong function of the particle Stokes number. A few experiments (Eaton 1994) suggest that attenuation is largest for Stokes numbers, based on the Kolmogorov time scale, of the order of 10 and particle diameters of the same order as the Kolmogorov length scale. Fundamental studies of turbulence modification in recent years have centered on the use of direct numerical simulation codes with the addition of point-force coupled particles or on the use of two-fluid models (Squires & Eaton 1990; Ahmed & Elgobashi 2000; Boivin *et al.* 1998). Both make major assumptions in their formulations that are not valid in the parameter regime noted above. Specifically, for the point-force coupling to apply, the particles must be much smaller than the grid scale. This cannot be achieved if the particle diameter is approximately the same as the Kolmogorov scale. In Reynolds averaged modeling, the most effort has been directed at correctly computing the so-called extra-dissipation due to particles in the kinetic energy transport equation. However, this term has been measured experimentally, and

it does not correlate well with observations of turbulence attenuation (Paris & Eaton 2001; Longmire & Khalitov 2002).

New experiments and or analyses are needed to cast light on the important phenomena that cause turbulence attenuation. In the experimental arena, measurements are needed that allow us to understand the scaling of turbulence attenuation with the various dimensionless parameters discussed above. Experiments should be conducted in simple turbulent flows such as grid turbulence, fully developed pipe or channel flow, or simple axisymmetric jets. Regardless of geometry, experiments must include a wide range of particle parameters in a single fixed facility. Ideally, the particle size would range from particles so small that they act as flow tracers and do not cause attenuation up to particles that are so large that they cause turbulence augmentation. (The experiments should also explore the effect of particle size distribution.) Flow parameters, especially the Reynolds number should be varied to assess the importance of the range of turbulent length scales in setting turbulence attenuation. Measurements should include at a minimum the mean and second order turbulent statistics for both phases, as well as particle/fluid velocity correlations.

More detailed experiments may be possible that cast light directly on mechanisms of turbulence modification. These would involve observations of the distortion of turbulent eddies in the neighborhood of particles. One of the biggest questions is how sub-Kolmogorov scale particles can affect energy containing eddies. Early thinking was that they only affected those eddies by collective action, similar to changes caused by an increase in viscosity. However, some recent results suggest that individual particles may substantially distort energy containing eddies (Eaton *et al.* 1999; Hishida & Sato 2001). This may be the key to understanding turbulence attenuation. Detailed simulations that resolve the flow around each particle may also contribute to our understanding of turbulence attenuation.

2.5 *Transition to turbulence*

The addition of particles and bubbles will modify instabilities such as the transition to turbulence that occur in a single-phase flow. Particles enhance the effective viscosity of a suspension and this may be expected to delay the transition to turbulence. On the other hand, particles create a disturbance in the fluid. Since the instability of single-phase pressure-driven channel flow is sub-critical and laminar planar Couette flow and pressure driven pipe flow are stable to infinitesimal disturbances for all Reynolds numbers, the finite amplitude disturbances produced by particles may play an important role in triggering a transition to turbulence. Particles may also modify the mean velocity profile and thereby influence flow transitions. An important challenge in performing experiments to test the effects of particles on the transition to turbulence is to identify diagnostics that can indicate the transition even in the presence of particles that cause fluid velocity fluctuations in a laminar flow.

3. Concentrated “fluid-like” suspensions

3.1 Derivation of equations of motion and constitutive equations

Although general conservation laws for the mass and momentum of each phase (or of the dispersed phase and the mixture) can be derived readily, these two-fluid model equations are not

closed. A simple starting place for considering the constitutive equations is to study the interaction of a single particle, drop, or bubble with the continuous phase. Much progress has been made at this level, whose validity is necessarily limited to very dilute systems. Even here, issues related to carrier phase turbulence (discussed above) remain unsolved.

A greater challenge concerns more concentrated suspensions where particle-particle interactions (both hydrodynamic interactions mediated by the continuous-phase fluid motion and direct collisions) play an important role. The understanding of the rheology of low Reynolds number suspensions has benefited greatly from consideration of simple model suspensions and simple flows for which analytical theories can be developed, tested, and then generalized to more complex systems. These studies often include direct comparison between theory, many-particle simulations, and ideal experiments specifically designed to test the theories. The time is ripe for applying a similar approach to multiphase flows where inertial effects are important on the particle length scale.

Theories have been developed for three special cases of inertial multiphase flow: rapid granular flows, gas-solid suspensions, and suspensions of high Reynolds number, nearly spherical bubbles (Lun *et al.* 1984; Sangani *et al.* 1996; Spelt & Sangani 1996). The latter two theories allow for relative motion of the continuous and disperse phases as well as shearing of the suspension. These theories are based on analogies with the kinetic theory of dense gases but allow for the dissipative effects of fluid viscosity and inelastic particle collisions. The theory of rapid granular flow assumes that particles undergo only instantaneous collisions (and not enduring solid-body contacts) and neglects all effects of the continuous phase. Theories of gas-solid suspensions have added the effect of a low Reynolds number gas, while assuming the particle Stokes number is large – a situation made possible by the large ratio of densities of the two phases (Sangani *et al.* 1996). Theories for bubble suspensions are limited to potential flow interactions among essentially spherical bubbles (Spelt & Sangani 1996). Although the simplest formulations of the theories consider nearly Maxwellian particle (or bubble) velocity distributions and Newtonian constitutive equations, more complex rheology including normal stress differences and viscoelastic responses can be captured by solving dynamic equations for all the components of the second moment tensor for the particle velocity distribution.

These theories have been validated extensively by comparison with many-particle numerical simulations based on the same physical assumptions as the theories (Sangani *et al.* 1996), but the comparison with experiment is quite limited. There is a need for careful experiments involving homogeneous flow to test the theories described above.

To develop a more general theory for disperse multiphase flows, a three-pronged strategy would be useful.

a) We should extend the theories for weakly dissipative multiphase flows to include stronger dissipative effects. For example, one can allow for finite Reynolds number effects in the theory for a gas-solid suspension by allowing for the enhanced viscous dissipation of particle kinetic energy at finite Reynolds number, including the Reynolds stresses of the gas, and considering the viscous force dipoles acting on the particles.
b) Theories for Stokes flow suspensions provide an opposite limiting case where viscous dissipation dominates over particle inertia. We should consider first the effects of fluid inertia on the rheology of a suspension at small, but non-zero Reynolds number, and establish a second beachhead in our attempt to understand moderately dissipative systems (Lin *et al.* 1970).

c) It is now possible to simulate the dynamics of particle or bubble suspensions (Bunner & Tryggvason 1999). Much of the earlier work has been aimed at developing the numerical methods and verifying the accuracy of their predictions for a given initial particle configuration. We should perform more extensive simulations with adequate time and/or ensemble averaging and a closer connection between simulation and averaged-equation modeling to reap the full benefit of these numerical simulation methods. The results of these simulations can be used to test approximate theories that are developed as extensions of the exact asymptotic results that can be obtained in the weak and strong dissipation limits.

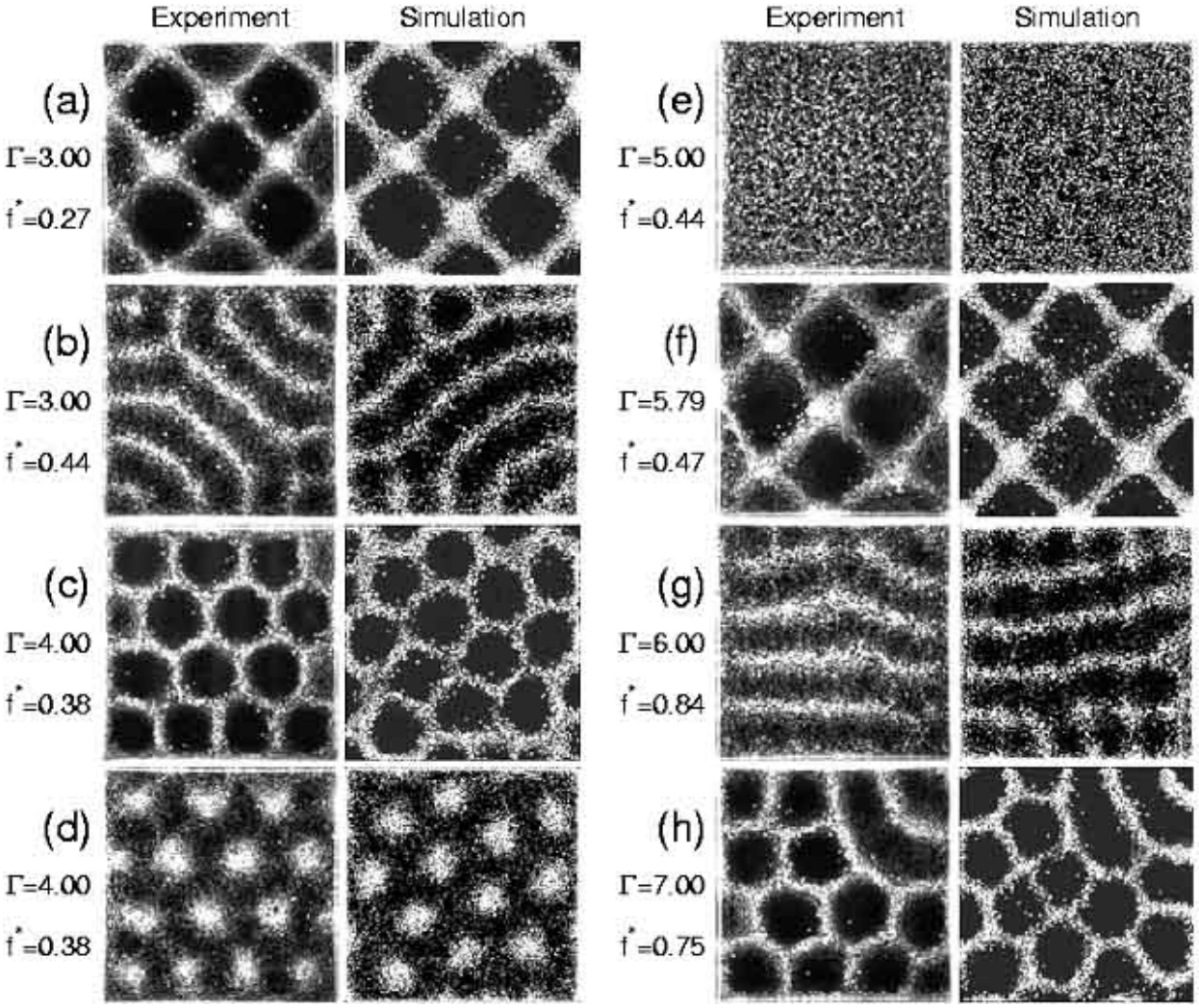

Fig. 3: Patterns obtained in a laboratory experiment and an event-driven particle dynamics simulation of a thin layer of particles in a vertically oscillated container, as a function of the container acceleration amplitude Γ (relative to the gravitational acceleration, g) and the non-dimensional frequency $f^* = \left(f\sqrt{h/g}\right)$ where f is the frequency and h is the height of the layer. Patterns 9a) – (e) oscillate at f/2, (f) – (h) at f/4. The layer was 5.4 particle diameter deep, and the brightness of the photographs indicates height of the layer. The experiment used lead particles (0.5 mm diameter). The same adjustable parameters in the simulation (restitution coefficient = 0.7 and coefficient of friction = 0.5) yield good agreement with experiment over the entire range of conditions shown, suggesting that simulations can capture the experimental results. Source: Reference (Bizon *et al.* 1998).

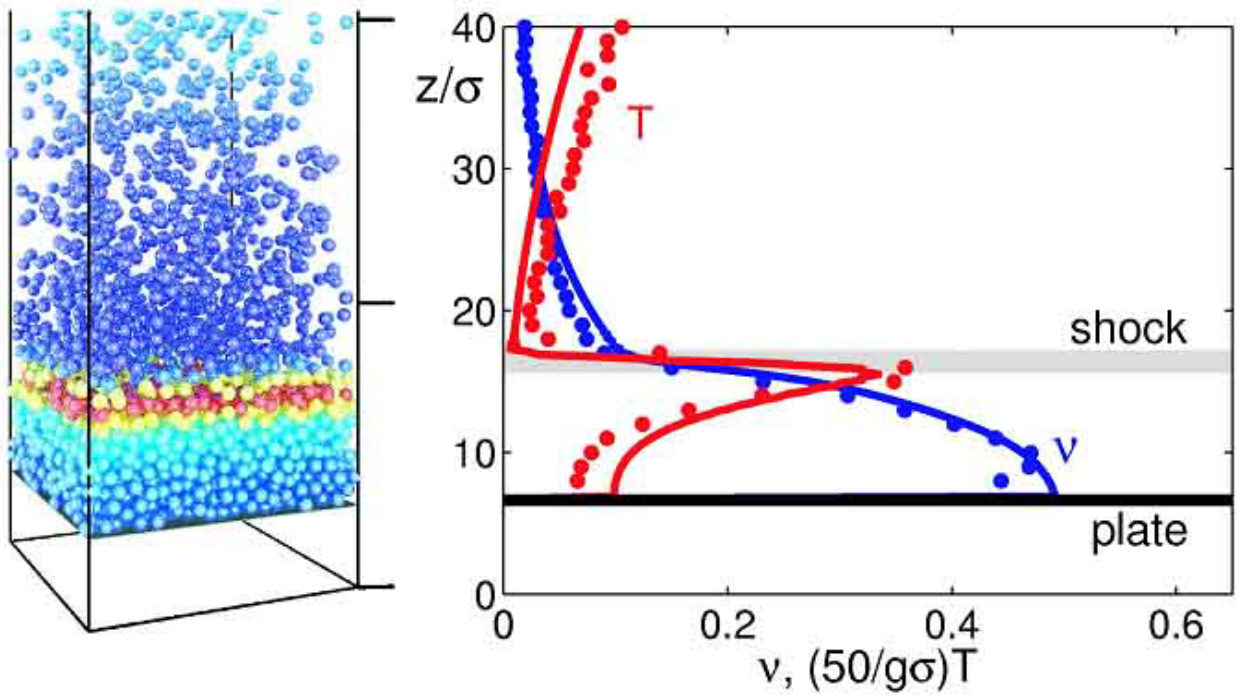

Fig. 4: The picture on the left shows results from a particle dynamics simulation of a granular layer in a vertically oscillating container, at a time that is 22% of a cycle past the time when the container was at its lowest point. The particles are color-coded according to the granular temperature, T: high T in red, low T in blue. The graph on the right compares results from the particles dynamics simulation (dots) and a simulation of continuum equations (continuity, momentum and granular energy balance equations) for the horizontally averaged volume fraction ν (blue) and temperature T (red), as functions of z (ordinate). The horizontal axis label is correct for ν; T is shown in units of gd/50, where d is particle diameter. The good agreement suggests that continuum equations are able to capture the simulations. Source: Reference (Bougie *et al.* 2002).

It is vital to test theory and simulation against physical experiments. Such comparisons are beginning to appear (Bizon *et al.* 1998; Bougie *et al.* 2002) for the case of rapid granular flows (see Figs. 3 and 4), but remain to be performed in multiphase flows. As discussed below, instabilities may occur in these suspensions on a length scale of the order of 10 – 50 particle diameters, so a narrow-gap channel or Couette, in which the gap thickness is only 5 – 10 particle diameters, may be necessary to achieve a stable flow. A good understanding of the boundary conditions on the averaged equations of motion is essential to achieve a quantitative comparison of theory and experiment. Boundary conditions for granular flows have received much attention (Hui *et al.* 1984; Jenkins 1992; Louge 1994), but the current understanding of boundary conditions in two-phase flows is more limited.

The shear flow of neutrally buoyant solid particles over a wide range of particle Reynolds numbers warrants careful study. A recent investigation (Hunt *et al.* 2002) shows that the classic experiment of Bagnold (Bagnold 1954) on high Reynolds number neutrally buoyant particle shear flow did not truly probe the effects of the particle phase on the suspension rheology.

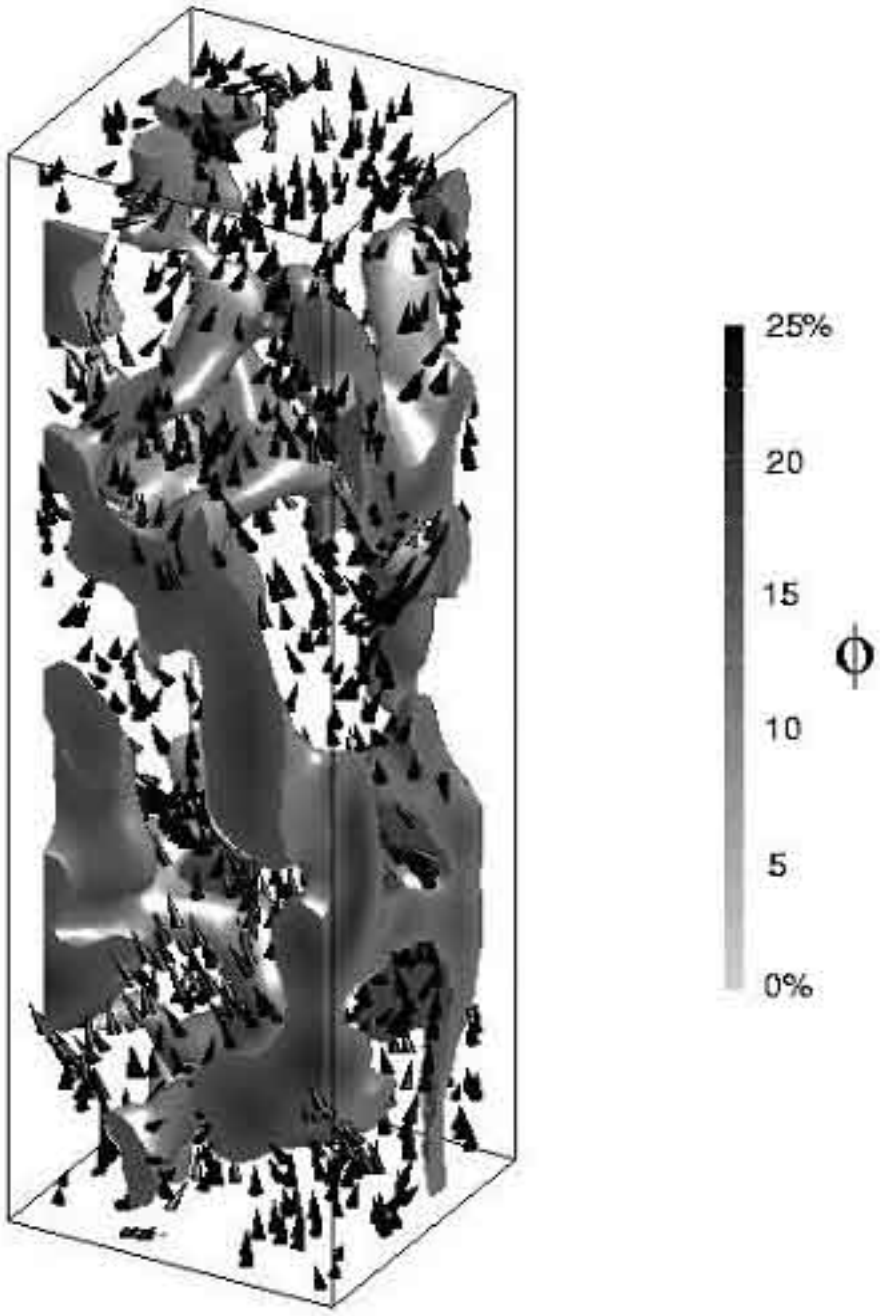

Fig. 5: Snapshot of solids volume fraction and gas velocity fields in a 3D simulation. Cones represent gas velocity, with orientation and length indicating direction and magnitude, respectively. A contour plot of the surface in the interior of the domain where particle volume fraction is 0.05 is shown. The solids volume fraction at points on the faces is as indicated in the grayscale (provided it exceeds 0.05). The mean solids volume fraction in the domain is 0.05. Simulations were performed in a periodic box using continuum balance equations. The mean pressure gradient in the vertical direction balanced the weight of the suspension per unit volume. An initially uniform state gave way to a time-dependent structure, and the figure shows an instantaneous snapshot of the time-dependent solution. Source: Reference: (Agrawal *et al.* 2001).

Present theories of multiphase flow typically do not incorporate effects of bubble or drop deformation. It is now possible to simulate interacting deformable bubbles (Esmaeeli & Tryggvason 1999), but we require a theoretical framework that accounts for bubble deformation. There is a mean bubble shape that is induced by the mean relative motion of the phases and the shear flow and this has a large effect on the drag, added mass and lift forces acting on the bubbles (Magnaudet & Eames 2000; Sankaranarayanan *et al.* 2002; Sankaranarayanan & Sundaresan 2002). However, there may also be important effects of bubble shape oscillations induced by the hydrodynamic disturbances caused by the bubbles and by bubble-bubble collisions.

It is well known that the dynamics of fluidized gas-solid suspensions can be altered profoundly by the addition of fines, which alter the particle size distribution (psd). Theories for the effect of psd on the rheological behavior of the fluid-solid suspension must be developed and validated.

3.2 *Instabilities of multiphase flows*

It is unusual to achieve a homogeneous, laminar multiphase flow when inertia is important on the particle or bubble length scale. The homogeneous state of a particle suspension translating relative to a gas or liquid phase (fluidized bed) or a bubble phase rising through a liquid (bubble column) is almost always unstable and gives way to non-uniform structures spanning a wide range of length and time scales (Lammers & Biesheuvel 1996; Agrawal *et al.* 2001; Sundaresan 2002). It is known that rapid shear flow of granular materials is unstable for sufficiently large gap thickness (Nott *et al.* 1999). Although stability analysis has not been performed for bubble suspension and gas-solid suspensions subject to simple shear flow, similar instabilities may be anticipated. Bidisperse suspensions also undergo instabilities that are driven by the interactions between the various particle species. It is likely that additional mechanisms of instability remain to be discovered.

To obtain a proper understanding of these instabilities, it would be valuable to directly extract evidence of the instabilities from particle-scale numerical simulations and compare the results directly with analysis of the averaged equations of motion. These comparisons should include testing theoretical predictions for the conditions of marginal stability and comparing simulated inhomogeneous flows to the solutions obtained from averaged equations (e.g., see study on sedimenting bidisperse gas-solid suspensions by Valiveti and Koch (Valiveti & Koch 1999)). Such comparisons will serve as stringent tests of the averaged equations.

In addition to understanding the marginal conditions for instability and having a mechanistic understanding of the instabilities, it is important to be able to model flow behavior of suspensions that are unstable. Usually, the typical length scale of the dominant mode through which a homogeneous state of a concentrated suspension loses stability is quite small (only a few particle or bubble diameters) and inhomogeneous suspension flows often manifest structures ranging from the size of the device down to the length scale of the dominant primary instability. First, the validity of the averaged equations when gradients arise on such short length scales is not clear. The averaged equations, as derived, are of limited value in solving many engineering problems involving large process vessels, as the resolution required to capture all the spatiotemporal structures is prohibitive (Sundaresan 2000). It would be desirable to develop more manageable models that are useful for probing macro-scale flow after averaging over both

the statistics associated with individual particle motion and the statistics of the unstable particulate flow structures (Agrawal *et al.* 2001; Zhang & Van der Heyden 2002). The latter averaging may be performed at all scales (analogous to the RANS approach in single phase flow) (Hrenya & Sinclair 1997) or over a limited scale as in large-eddy simulations. One strategy for developing such models is to obtain solutions of the averaged equations of motion (obtained by averaging over the statistics associated with individual particle motion) for unstable situations and gather the requisite statistics (Agrawal *et al.* 2001). (See Fig. 5.) Only a few computations of his kind have been performed so far, and they suggest that fluctuations associated with the unstable structures very quickly overwhelm those at the level of the individual particles (Agrawal *et al.* 2001), so accounting for the former is crucial. It should be noted, however, that the validity of the averaged equations in such calculations is not clear, as sharp gradients will be on short length scales, which are invariably not considered in the formulation of the averaged equations. Therefore, it is important to validate this approach by comparing with particle-scale simulations (which would be restricted to smaller flow domains) and experiments.

Very little is known about the fluctuation statistics in such unstable flows. It appears that, to a large extent, the energy is extracted from mean flow by inter-phase drag, giving rise to small-scale structures, which interact with each other yielding larger scale fluctuations. Unlike single phase flow, macro-scale shear plays only a secondary role in these multiphase flows (Agrawal *et al.* 2001). Indeed, there is little reason to believe that the models for unresolved turbulence developed for single phase flow are even qualitatively correct for this class of multiphase flow problems. It now appears that coarse-grid simulations of multiphase flows should include the effects of unresolved structures via time-averaged and stochastic forcing terms (Loezos & Sundaresan 2001). Physical and computational experiments probing the fluctuation statistics are needed. Physical experiments involving intrusive measurements may end up perturbing the local instability associated with the relative motion between the continuous and dispersed phases. Nevertheless, such measurements may yield useful information on structures arising from instabilities that were initiated elsewhere in the vessel and transported to the probe location (Zenit *et al.* 2001). Non-intrusive measurement techniques, which can provide sufficiently fine spatial and temporal resolutions, should be developed and deployed.

4. Dense "solid-like" particulate systems

An important limitation of the theories noted above is that they deal only with "fluid-like" suspensions where the particles interact through the interstitial fluid and ephemeral impulsive interactions. It is important to understand the conditions leading to transition from fluid to solid like behavior, where the particles interact with each other through enduring contact between each other and boundaries, and how they depend on particle volume fraction, frictional properties of the surfaces involved, cohesive interparticle forces, dissipative collisions, etc. Such transitions are known to play a dominant role in the behavior of many granular materials and have been observed in fluidized beds. For example, defluidization experiments indicate that particle assemblies pack in a solid-like state over a range of volume fraction (ϕ_{min}, ϕ_{max}) (Tsinontides & Jackson 1993; Valverde *et al.* 1998). Packing at higher concentrations can be achieved only by means such as tapping, flow on-off toggles or large imposed stresses. Compressive and dilational yield stress characteristics of the assemblies (at various solids volume fractions) dictate the dynamics of the granular phase in this solid-like state. Theories for these stresses based on particle-level characteristics such as cohesive interparticle forces, size

and shape and their distributions, frictional contact between particles, etc. must be developed, so that one can understand how to manipulate flow behavior by tailoring particle characteristics. Particle-scale simulations can shed light on conditions leading to the fluid-solid transition and the network of force chains in a solid-like state.

Our knowledge of frictional stresses in flowing granular assemblies is primitive. Quantitative modeling of the performance of many devices such as fluidized and spouted beds, standpipes, dense phase pneumatic conveying and mixing equipment, *etc*. require an improved knowledge of frictional stresses. While constitutive models for quasi-static flows developed in the soil mechanics field may be good starting point, there is growing evidence that these must be modified to bring in the effect of strain rate fluctuations to transition region between quasi-static and rapid flow regimes (Savage 1998). These should be tested with simulations and experiments.

4.1 *Interaction of time scales*

Transition between fluid-like to solid-like behaviors is determined by the stress relaxation time in many rheological theories for polymeric materials. While spatial correlations in dense granular flows have received much attention, relatively little has been done on temporal correlations. The magnitude of the slowest relaxation time associated with the temporal correlations of particle interactions in dense particulate systems relative to the hydrodynamic time scale is an important quantity, which determines whether viscoelastic models are needed to describe the suspension rheology or not (Zhang & Rauenzahn 2002). The circumstances (in terms of time scales) where viscoelastic models for suspensions are essential to describe the rheology properly and the structure of such models are important frontiers.

4.2 *Role of particle spin and frictional interactions with boundaries*

The interaction of particles with each other and with solid boundaries involves friction and particle spin (Bizon *et al.* 1998; Jenkins 1992; Louge 1994; Jenkins & Louge 1997). Formal theories predicting the transport of particle angular momentum near boundaries are beginning to emerge (Hayakawa 2002). These should be extended to gas-solid flows and tested with simulations and experiments.

Solid friction is another topic that remains largely a mystery. For example, in granular flows, friction can vary with relative contact speed; it can also be hysteretic or intermittent (e.g., stick-slip). Its understanding is crucial to predict the interaction force that is tangent to a solid surface. More globally, it is important to distinguish whether particles are engaged in long-lasting frictional contact with boundaries or more ephemeral impulsive interactions. This distinction is not only crucial to gravity-driven granular flows (Louge & Keast 2001), but also to any gas-solid flow where particles remain in contact with walls (Griffith & Louge 1998).

4.3 *Mixing and segregation*

In our discussion thus far, we were primarily concerned with flow. However, there are many systems where the mixing and segregation resulting from flow take on a greater importance than the flow itself. These arise in dry granular materials (particles surrounded by air), wet granular materials (particles in a humid environment or coated with a small amount of fluid) and slurries (dense particles completely immersed in a lighter liquid). Most common

applications involve *slow flows*, where particles roll past each other and are in contact with several neighbors at the same time. This is relevant to tumbling, heaping, motions induced by moving objects, such as blades, etc. Both non-cohesive and cohesive systems are of interest. The most interesting and practically important case corresponds to bimodal populations, where the particle variation may occur in size, shape, density or surface properties.

Granular materials segregate. Small differences in size, density, surface roughness, or particle shape lead to flow-induced segregation (Ottino & Khakhar 2000). This is a complex and imperfectly understood phenomenon. Segregation issues are, however, unavoidable in practice. Fig. 6 highlights the dramatic changes in the mixing-segregation structures caused by small changes in the operating conditions, in this case the degree of filling in a tumbler. To describe segregation, we not only need models for flow, but also constitutive models for segregation fluxes under various flow states. The bulk of the studies in the literature consider only two special cases: a mixture of particles of the same density and shape, but two different sizes, and a mixture of particles of same size and shape, but different densities. Models for combined size and density segregation remain to be developed. An even more complicated issue is particle shape. Virtually nothing is known about how to incorporate the role of cohesion. This area needs significant investments in theory and controlled experiments. In turn, experiments depend in significant advances in non-invasive techniques.

A great deal of segregation modeling work is based on particle dynamics (PD) simulations which require precise physical properties (Young modulus, restitution coefficients, Poisson ratios, etc.) (Cleary *et al.* 1998). Even in the case of spheres there are many important questions that need to be clarified.

- What is the parametric sensitivity of the results to the various parameters in PD models?
- How should one handle walls? How sensitive are the results to this choice?
- How can one match a real material – non-spherical, multi-sized – to input parameters?

The PD simulations can typically handle about 10^4-10^5 particles (Shinbrot & Muzzio 2000), in contrast to practical systems which have $>> 10^9$ particles and invariably require continuum modeling approach. Development of multi-scale models involving continuum and PD approaches to handle macro- and meso- scale phenomena is a scientific challenge that can be tackled within the next 5 – 10 years.

Fig. 6: Experimental results on quasi two-dimensional tumbling mixers with glass spheres of different sizes (0.8 mm blue, 1.2 mm clear, and 2.0 mm red). All three systems started well mixed and everything is equal except the degree of filling. From left to right: less than 1/2 full, 1/2 full, more than 1/ 2 full.

A possible fruitful area is control of flow and especially, mixing and segregation, via manipulation of the cohesive force. A long-range goal is to identify suitable parameters that will indicate when cohesion may either mitigate or enhance the possibility of achieving a segregated distribution of particles.

The understanding of the competition between mixing and segregation is in its infancy. All studies to date are restricted to 2 dimensions and even in the classical example of axial segregation there is no agreement as to the underlying governing mechanism. A fruitful area for the next 5 years is the study of 3D systems. This area needs significant investments in theory and controlled experiments. Again, experiments depend in significant advances in non-invasive techniques.

It is apparent that our ability to characterize granular flows is hindered by a lack of suitable experimental techniques. The main difficulty is that granular materials are opaque; this prevents the use of many common methods that have been successful in fluid flows. Recent developments in Magnetic Resonance Imaging (Nakagawa *et al.* 1993), Particle Image Velocimetry (Lueptow *et al.* 2000), Particle Tracking Velocimetry (Jain *et al.* 2002), X-ray imaging (Harwood *et al.* 1973) and Positron Emission Particle Tracking (PEPT) (Parker *et al.* 1997) offer tremendous opportunities in the next decade. These should be applied to the precise characterization of building block flows, such as shear layers.

The goal of all the techniques above is to obtain fields, one aspect being how to construct a velocity field for the flow. Two main issues are unavoidable: The relatively low spatial resolution of the experimental measurements and the fact that for many experimental techniques (e.g. PEPT) significant statistics are only obtained through several different experimental measurement series. Techniques must be developed to reconstruct the "true" velocity field. In particular one has to consider the possibility of very irregular, if not chaotic, flows. Such flows may not be easily distinguished in the coarse-grained velocity fields available from the experimental data.

Scale up of granular processes is notoriously difficult and there is little guidance at the present time. However, flows in granular materials are often restricted to thin regions of rapid surface flow with the rest of the material suffering only slow plastic re-arrangements. This seemingly trivial observation leads to understanding of the entire system and to the concept that an understanding of surface flows constitutes the key element for scale up of granular flow processes. However, before this becomes possible, shear flows in tumbler and heap formation need to be thoroughly characterized. The goal is to predict layer thickness and shear rates based on first principles.

5. Concluding remarks

Disperse flows are encountered in a variety of contexts: industries dealing with energy, pharmaceuticals, bulk and specialty chemicals, and consumer products, agriculture, transportation and conveying, and space exploration. They are studied, researched and applied in many academic disciplines including engineering (chemical, civil, environmental, materials science, mechanical and metallurgy), physics, atmospheric sciences (climate change, meteorology) and geosciences. Progress in this field impacts a broad range of industries and academic disciplines.

The set of challenges outlined in this report is by no means complete. It is simply a collection of some important problems which can realistically be tackled in the next decade. Resolution of these issues will lead to significant advances in our understanding and modeling capabilities.

Although this report may appear to be slightly tilted towards theory, the need for experiments and simulations cannot be over-emphasized. Model experiments which can be used to guide the development of theory and its validation are extremely important. By and large, experiments, theory and simulations are done by different research groups. It is desirable that these groups seek alliances quite early on so that experiments to probe specific aspects of theory and/or simulations will also be identified and performed.

In this report, we have outlined a number of scientific challenges which represent building blocks for comprehensive understanding and modeling of disperse flow encountered in a variety of technologies and in nature. These building blocks range from flow characteristics at the length scale of individual particles or bubbles and contact mechanics to computational and physical experiments aimed at validating models for specific aspects of multiphase physics. The importance of convergent, integrative studies which will bring together these building blocks cannot be understated. At the same time, it is important to recognize that research on building blocks and their validation studies (such as those described in Figs. 2 and 3) need not (and should not be required to) directly address specific technological applications. Research on building blocks will be divergent, often raising more questions than answers. It is hoped that the present roadmap communicates to the reader the importance of merging integrative and divergent studies. The integrative studies are essential to understand how processes occurring over a broad range of length and time scales interact to influence multiphase flow on a large scale, while studies on building blocks will expose and resolve deficiencies in our fundamental understanding of multiphase flow fundamentals.

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WORKSHOP ON SCIENTIFIC ISSUES
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APPENDIX 3

REPORT OF STUDY GROUP ON COMPUTATIONAL PHYSICS

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APPENDIX 3

REPORT OF STUDY GROUP ON COMPUTATIONAL PHYSICS

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1. Introduction

Since the early days at Los Alamos National Laboratory, four decades ago, theoretical work in multi-phase flow has relied on computation. This trend has undergone a marked increase in the last few years, thanks to major developments in algorithms and computer power. As a consequence, the prediction of remarkable progress in this area in the coming few years is an easy one to make.

Computations of multiphase systems play many roles. First and foremost is the generation of basic understanding of the behavior of well-defined systems, not only through the simulation of the actual physical process, but also with the aid of computational "experiments." Multiphase flows are notorious for the difficulties in setting up fully controlled physical experiments. However, computationally, it is possible, for example, to include or neglect gravity, account for the effects of a well-characterized surfactant, and others. It is now possible to compute routinely the behavior of relatively simple systems, such as the capillary breakup of jets and the shape of bubbles. The next few years are likely to result in an explosion of results for such relatively simple systems where computations will help us gain a very complete picture of the relevant physics over a large range of parameters.

At the opposite end, one finds the great complexity of scales and phenomena of practical multiphase flows, where the behavior of the system is often exceedingly intricate (e.g., churn-turbulent, developing gas-liquid flows) or affected, at the large scales, by small-scale processes (e.g., nucleation, wetting, contact line motion, and others). The simulation of phenomena of the latter type must rely on effective computational methods for multiscale problems, which are still in their infancy. At a practical, industrial level, the simulation of phenomena of the former type must rely on an averaged description and closure models to account for the unresolved

phenomena. In this sense, the situation here is similar to single-phase turbulent flows where, in the last two decades, simulations have played a major role, e.g., in developing large-eddy models. Simulations are starting to play a similar role for multiphase flow and it is already clear that the opportunities for major progress are enormous.

Next to the problem of dealing with scale integration, it is a remarkable development that the next challenge is perhaps not so much increasing the computational power as understanding how to best exploit the enormous amount of data generated by simulations. Our very thinking patterns, developed in an environment where analytical and experimental results were limited, are becoming inadequate tools to deal with the newfound abundance of data. It is of the utmost importance that progress in computation be paralleled by new theoretical frameworks that make sense of and condense the results. ***Computation has made theory more relevant.***

The work of Magnaudet & Eames (2000) on the flow around a spherical bubble and that by Bagchi & Balachandar (2002) around spherical particles are typical examples of the role of computation in answering fundamental questions in multiphase flows. While these activities will undoubtedly increase, it must be recognized that such simulations are, in some sense, relatively straightforward extensions of what is currently possible. It is in the examination of very complex, very large-scale systems, where it is necessary to follow the complex evolution of an enormous range of scales for a long time, that the major challenges and opportunities lie. Such simulations, in which it is possible to get access to the complete data and to control accurately every aspect of the system, will not only revolutionize our predictive capability, but also open up new opportunities for controlling the behavior of such systems.

At the same time, the new insight offered by detailed simulations will be instrumental in developing better closures for averaged-equations models. While, for the foreseeable future, it is hard to imagine another form for practical engineering simulation tools, it must be recognized that the classical two- (or multi-) fluid models suffer from several ills as well as limitations. Major new advances are envisaged which will capitalize on numerical capabilities to lead to farther-reaching modeling tools suitable for practical applications.

2. Numerical methods

Direct numerical simulations of multiphase flow, where the full continuum equations are solved on a computational grid sufficiently fine to resolve all continuum scales, date back to the origin of computational fluid dynamics at Los Alamos in the early and mid sixties. The difficulty of following the deformation of an unsteady fluid interface separating phases of different properties, and limited computer power restricted the complexity of the systems that could be examined. During the last decade, however, major progress has been made, using a variety of numerical techniques. Before we discuss direct numerical simulations of multiphase flows (next section), here we will briefly review the methods that have been used for such simulations.

The oldest and still the most popular approach to compute multifluid and multiphase flows is to imbed the front directly on a regular, stationary grid. The Marker-And-Cell (MAC) method, where marker particles are used to identify each fluid, and the Volume-Of-Fluid (VOF) method, where a marker function is used, are the best-known examples. Traditionally, the main difficulty in using these methods has been the maintenance of a sharp boundary between the different fluids and the computation of the surface tension. A number of recent developments, including a technique to include surface tension developed by Brackbill, Kothe *et al.* (1992) and

the use of "level sets" (see, e.g., Sussman *et al.* 1994) to mark the fluid interface have increased the accuracy and, therefore, the applicability of this approach. A review of the VOF method can be found in Scardovelli & Zaleski (1999). The level set method is reviewed by Osher & Fedkiw (2001) and by Sethian (2001). Recent additions to the collection of methods that capture fluid interfaces on a fixed grid include the CIP method of Yabe (1997) and the phase field method of Jacqmin (1999). For reviews see Yabe *et al.* (2001) and Jamet *et al.* (2001). In these methods, one set of equations is used for the whole flow field and the various material properties form fields that change discontinuously across a phase boundary. Surface terms therefore have to be added as singularities at the interface. This "one fluid" approach has also been used by Tryggvason and collaborators (Unverdi & Tryggvason 1992 and Tryggvason 2001) to successfully simulate a number of multiphase systems. The difference between Tryggvason's approach and the "front-capturing" methods listed above is in the use of explicit marker points to follow the fluid interface. Since the one field approach is retained (unlike front tracking methods where each fluid is treated separately) the method is best described as a hybrid between front capturing and front tracking. The fictitious domain method of Glowinski *et al.* (2001), where solid body motion is enforced by Lagrangian multipliers also falls into this category.

The second class of methods, and the one that offers the potentially highest accuracy, uses separate, boundary fitted grids for each phase. The steady rise of buoyant, deformable, axisymmetric bubbles was simulated by Ryskin & Leal (1984) using this method. Several two-dimensional and axisymmetric computations of both the steady and the unsteady motion of one or two fluid particles or free surfaces can be found in the literature. This method is best suited for relatively simple geometries, and applications to complex fully three-dimensional problems with unsteady deforming phase boundaries are very rare. The simulation of a single unsteady three-dimensional bubble by Takagi & Matsumoto (1994) is, perhaps, the most impressive example.

The third class is Lagrangian methods where the grid follows the fluid. Examples of this approach include the simulations of the unsteady two-dimensional motion of several particles by Feng *et al.* (1994, 1995) and Hu (1996) and axisymmetric computations of the collision of a single drop with a wall by Fukai *et al.* (1995). While this appears to be a fairly complex approach, Johnson & Tezduyar (1997) and Hu *et al.* (2001) have recently produced very impressive results for the three-dimensional unsteady motion of many spherical particles.

The fourth category is front tracking where a separate front marks the interface but a fixed grid, only modified near the front to make a grid line follow the interface, is used for the fluid within each phase. The main developer of this approach has been Glimm and collaborators (see Glimm *et al.* 2001).

In addition to front tracking methods that are, in principle, applicable to the full Navier Stokes equations, specialized boundary integral methods have been used for both inviscid and Stokes flows. For a review of Stokes flow computations, see Pozrikidis (2001) and, for a review of computations of inviscid flows, see Hou *et al.* (2001). The most recent addition to the collection of methods capable of simulating multiphase flows is the Lattice Boltzman Method (LBM) reviewed, for example, by Chen & Doolen (1998) and by Sankaranarayanan *et al.* (2002).

Because many of these methods have been developed relatively recently, no clear "winner" has yet emerged. Indeed, it is likely that, for each method, there is a set of problems where it exhibits advantages over others. Furthermore, it is equally likely that often the "best" method is the one that the investigator is most proficient with.

While numerical methods can be made more efficient, more accurate, and more robust, the most pressing need in the next decade is perhaps not in the development of new methods but in the use of the available methods to advance our understanding—to formulate a **theory** of dispersed flows.

3. Complexity

Multiphase flows are inherently complex. Even limiting oneself to the relatively 'simple' case of disperse flows, as soon as realistic numbers of particles and turbulence are involved, the possibility of carrying out DNS quickly dissolves. Just as in single-phase flow, large-eddy simulation appears to be a natural way to attack this situation. This is an important topic that so far has been scarcely studied, but which needs to be pursued. It seems likely that there should be a unifying formulation that allows one to start with the full equations (DNS) and naturally progress toward averaged equations with point particles and extended particles in between. Some highly non-trivial flows could be modeled if large-eddy simulation of disperse flows were developed. For example, in annular flow, one may envisage an LES model to describe the flow of the core gas with suspended droplets, and a fully resolved simulation of the interface between the gas core and the liquid film.

More generally, the complexity of multiphase flow requires a reduced description which is—and most likely will remain—embodied in a set of averaged equations which, in order to be realistic and reliable, must be greatly improved with respect to the models currently in existence. The efforts of the past several decades have shown that it is futile to hope to attain this goal solely on the basis of experiment and 'simple' fluid dynamics (e.g., single particles in idealized situations). For example, the issues of ill-posedness of these equations and implications about their basic physical content continue to linger. The new and critical capability which now can be brought to bear on the essential task of improving these models is the recently developed ability to conduct non-trivial numerical simulations of relatively complex flows. It must be stressed, however, that while this point is obvious, the details of how to use simulations as a guide for the formulation of realistic and robust averaged-equations models are still very unclear. It is necessary and urgent that the passage from computational results to theory development be addressed as a specific problem in its own right.

While in some cases, such as the disperse flows mentioned above, complexity of scales can hopefully be dealt with by means of reduced descriptions, there are others where such descriptions are not—or, at any rate, not yet—possible. The pinch-off of a liquid thread, the coalescence of bubbles or drops, the motion of a contact line are superficially simple examples where the full simulation requires the ability to simultaneously account for spatial scales ranging over some seven orders of magnitude. Another example, enhanced oil recovery, depends on phenomena ranging from the level of single pores (micron scale) to reservoir scale (kilometers). Dealing with such problems requires the development of new theoretical tools and computational procedures.

But complexity does not arise only from the interaction of different scales: the physics governing important phenomena can itself be complex. Even limiting oneself to the restricted class of the flow of two immiscible fluids, it is quite easy to develop a long list of situations much more complex than those considered in the previous section. If one looks beyond disperse flows, the complexity of the problems vastly increases, and so does the potential of

computational physics in attacking them. For example, no attempt has been made to carry out simulations of flow regime transitions. Churn-turbulent flow is a completely virgin territory. Phase distribution, the formation, evolution, stability, and break-up of slugs, and similar problems have only very recently begun to be addressed.

In a large number of engineering applications that involve multiphase flow, it is necessary to account for phase change, both between liquid and solid as well as liquid and vapor. Most materials used for man-made artifacts are processed as liquids at some stage, for example, and the way solidification takes place generally has major impact on the properties of the final product. The formation of microstructures, where some parts of the melt solidify faster than others, or solidify with different composition as in the case of binary alloys, is particularly important since the size and composition of the microstructure impact the hardness and ductility, for example, of the final product. Boiling is one of the most efficient ways of removing heat from a solid surface and it is therefore commonly used in energy generation and refrigeration, for example. The large volume change and the high temperatures involved can make the consequences of design or operational errors catastrophical and accurate predictions are highly desirable. The change of phase from liquid to vapor and vice-versa usually takes place in a highly unsteady manner, within thin diffusion layers and in the presence of very convoluted phase boundaries. Only a few examples of direct numerical simulations of both the effect of flow on the formation of microstructures during solidification and boiling have been published in the last few years (Beckermann *et al.* 1999; Tonhardt & Amberg 1998; Juric & Tryggvason 1998; Son & Dir 1998; Shin & Juric 2000; Esmaeeli & Tryggvason 2002) and this is likely to become a very active area in the next few years.

Other systems include more complex physics such as rheological effects (non-Newtonian fluids, polymer solutions), chemical reactions (e.g., combustion), three-phase systems (contact lines, liquid-gas-solid suspensions), thin films (boiling crisis, coalescence, break-up), electric and magnetic fields, and others. The potential for complexity is virtually unlimited and even relatively simple systems will put considerable demand on computational resources and the solution methodology. Thus, for example, the effect of electric fields on the boiling of binary mixture requires the solution of the fluid flow, species conservation, the energy equation and phase change, as well as an equation for the electric field. While pioneering work has been done on some of these problems, for the most part the field is wide open.

4. Conclusions

Computation serves many essential roles:

As a tool to develop our understanding of the basic physics: asking “what if” questions, clarifying the importance of physical effects (e.g., gravity, surface tension) by adding or removing them at will, and others.

As an aid in closing the averaged equations: just as the effectiveness and physical realism of different single-phase LES formulations can be judged by comparison with DNS results, closure relations can be developed and tested against numerical simulations.

As a means of solving actual problems: some relatively small-scale problems (e.g., in microfluidics) can be attacked by DNS; bigger problems can be treated by means of reduced formulations, such as averaged equations.

As a device to *learn to compute better*: it was argued before that it is necessary to develop new ways to deal with the large amount of data made available by the simulations. It is also necessary to learn how to ask more penetrating questions, how to develop more powerful algorithms, how to deal with problems having a multiplicity of scales.

As always, what is feasible is not necessarily interesting, and what is important is not necessarily feasible. While it is a trivial statement that it is at the intersection of the feasible and the important that real progress will be made, it is not always obvious where this intersection lies. In particular, computational research is often accused of generating vast amounts of trivial or unnecessary information. It is evident that the multiphase flow community must resist the temptation of using existing codes to generate yet another unnecessary paper, and focus instead on what is truly important and innovative.

The coming of age of powerful computational capabilities must be ranked as one of the most important turning points in the history of multiphase flow research. The last decade has seen the development of several extremely effective algorithms which, coupled with hardware of unprecedented power, make the computation of complex flows now possible. Although our abilities to directly simulate more and more complex multiphase systems will certainly increase dramatically in the next few years, it is important to realize that our desire to compute will always be ahead of our ability. Even if we could fully compute the behavior of a system, we may easily imagine, for example, that we might want to be able to incorporate simulations into a real-time control system that dynamically explored the consequences of several possible control actions. Thus, the condensation of knowledge obtained by direct numerical simulations into reduced or averaged models that allow faster predictions will remain at the core multiphase flow research for a long time to come.

Computing is linked to Mathematics, on one side, and to Computer Science, on the other. The first link has always played an important role in multiphase flow simulation, but the same cannot be said of the second. Fields where major progress is likely include visualization, data mining, programming philosophy and techniques, and others.

Before concluding, it is important to mention problems of a different nature, which have begun to emerge in all their seriousness in the last few years, namely the **education** of students and new researchers in the field of computation and how "computational knowledge" and even software is shared.

No experimentalist would build his/her tools completely from scratch; any experimental setup contains vital components, such as cameras and lasers, that are purchased from commercial vendors. With a few important exceptions, in this respect, the computational researcher is still operating in the dark ages. Although commercial codes are available for solving engineering problems, these codes are generally unsuitable for state-of-the-art research. Their limitations—such as robustness in favor of accuracy and limited access to the "guts" of the codes—are understandable, but the result is that most research codes must be written by the group that intends to use it. In many cases, a sophisticated piece of software does not survive the graduation of the student who wrote it. While many research groups make their codes available, and advanced codes have been turned into generally available packages, it is clear that a major cultural change is called for.

The complexity of developing fully parallelized software to solve the continuum equations (fluid flow, mass and heat transfer, etc), where three-dimensional interfaces must be handled and the grids must by dynamically adapted, are putting such simulations beyond the domain of what a typical Ph.D. student, or even a small research team, can accomplish over the

span of a few years. To make the learning and development process faster it is essential to find ways to provide adaptable, well-documented, software components to new users. A related problem, which plagues the developers of new codes, is their validation; the complexity of these codes is such that in many cases the only tools for validation are other, independently developed, codes. In this situation, the ability to validate a code often depends on the circle of acquaintances of the developer—obviously an unacceptable situation. And, finally, integration: one can envisage a future in which, in some cases, complex computational research tools will be assembled from components developed by different research teams. How is this going to happen? How will this affect the perceived "productivity" of the individual teams? How do we move from a "cottage industry" to a "big science" mentality? The major onus in establishing a climate in which these questions will find a good answer rests on the funding agencies.

5. Overview of the individual contributions

In addition to the authors of this Summary, the task group on Computational Physics consisted of S. Balachandar (University of Illinois at Urbana), Shiyi Chen (Johns Hopkins University), Lance Collins (Cornell University), Martin Maxey (Brown University), Olivier Simonin (University of Toulouse), and Theo Theofanous, T.N. Dinh, and R.R. Nourgaliev (University of California at Santa Barbara). The documents they prepared will be found in a special issue of *Multiphase Science and Technology* and in the Proceedings of the Workshop on Scientific Issues in Multiphase Flow.

Rather than focusing on these individual contributions, in the previous pages we have tried to outline the broad issues emerging from them as well as the discussions held at the Workshop. Here we present a brief synopsis of each participant's input to the task group.

• Balachandar addresses the hydrodynamic forces exerted on a single rigid particle. He points out the difficulty in unambiguously prescribing the fluid force when the particle is immersed in a complex, unsteady, and spatially-dependent flow. The conclusion that the reader draws from this survey is that point-particle Lagrangian models, in which the motion of each point particle is dependent on the specification of the fluid force in terms of local flow parameters, are perhaps near the end of their useful life. It is necessary to go beyond these models and develop more realistic extended-particle models and computational techniques. This point is also made in several other contributions (Collins, Maxey, Tryggvason).

• Chen gives an overview of lattice Boltzmann methods. He points out the great computational advantages (efficiency, flexibility, ease of parallelization, applicability to high-Knudsen number regimes, and others), but also some drawbacks (e.g., the inability to easily handle large density ratios between the phases). The achievements of lattice Boltzmann methods are impressive and it is useful to continue work in this area.

• Collins focuses on the turbulent transport of aerosol particles, turbulent modulation by a suspended phase, collision, coalescence, and break-up of drops and bubbles, and the effect of polymers on turbulence. While cautious on the wisdom of continuing work on point-particle models, in the systems he considers (aerosol transport, cloud physics, inhalation drug therapy, polymer solutions, and others) particles are indeed very small and dilute and these models are still useful. They must be supplemented, however, by progress on basic issues such as collision and coalescence modeling. Like Balachandar, he touches upon the difficulties associated with an understanding of turbulence modulation in the context of these models. He stresses the need to

tackle problems in a concerted way by theory, computation and experiment, all addressing the same parameter range, so as to leave as few “loose ends” as possible.

• Hu presents an exhaustive review of the finite-element methods available for the description of fluid-particle flows: arbitrary Eulerian-Lagrangian scheme, stabilized space-time method, distributed Lagrange multiplier. A common problem is the description of particle collisions, which are numerous and frequent in dense systems: a first-principles description seems impractical, due to computational time and to the need to include features such as particle roughness. Hence approximate methods to deal with collisions are necessary, but not yet adequately developed. Hu also presents several computational examples of particles suspended in Newtonian and visco-elastic fluids.

• Maxey's contribution recognizes the progress made with point-particle models, but stresses the importance of moving beyond them for further progress. He describes in detail the Force Coupling Method for the Lagrangian tracking of finite-size particles suspended in a fluid and considers it as a “bridge” between point and extended particle models. While one should not lose sight of the many challenges which exist at the level of computational techniques and their efficient implementation, he stresses that it is equally important to put the computational results to good use for an understanding of the physics and the development of approximate models of engineering value. This is a crucial point also stressed by Tryggvason and Prosperetti.

• Prosperetti treats several issues related to the averaged description of multi-phase flow: averaging, stability, hyperbolicy, and the essential role of direct numerical simulation in guiding the formulation of more satisfactory averaged equations. He makes the point that averaged equations must necessarily be the workhorse of theoretical and design work involving multiphase flows in engineering, and discusses some basic limitations of the available equations. In this perspective, the development of reliable equation models can be seen as the over-arching goal of multiphase flow research. In his view, the direct numerical simulations newly made possible by progress in algorithms, software and hardware are the key ingredients in developing a better generation of averaged equations.

• While Simonin feels that point-particle Lagrangian models need to be improved, he is optimistic for their life in the near future: (1) Dynamic effects in particle-laden turbulent flows have been studied extensively for the last 15 years, but many open questions of great interest remain concerning, for example, heat and mass transfer in reactive turbulent flows or particle-turbulence, particle-particle and particle-wall interactions for non-ideal hard-sphere particles; (2) In a short-term perspective, LES is the only approach which allows one to represent realistic turbulent Reynolds number and to account for complex geometries. LES combined with a Lagrangian approach is well suited for intermediate-scale numerical simulations provided it is coupled with accurate true DNS for the computation or modeling of the subgrid effects. Furthermore, he argues that an LES methodology should be developed for the two-fluid equation models.

• Theofanous, Dinh, and Nourgaliev make a strong case that the multiscale treatment of multifluid flow holds the key to progress. They describe an approach that decomposes the simulation of a topologically complex multiphase flow into simulation of large-scale discontinuities and solution of disperse flow problems in domains dynamically encapsulated by such discontinuities. They consider these multi-scales approaches as the first step toward uncovering the principles that govern pattern formation in multiphase flow. In the face of the huge variety of multiphase flow phenomena, they advocate a strategy in which certain prototypical problems should be worked on first, so that the lessons learned in this process can

fruitfully be applied to other cases. Their list of these “higher priority"’ problems includes slug flow in horizontal and inclined pipes, churn-turbulent flow in large-diameter pipes, and critical heat flux in pool and flow boiling.

• Tryggvason describes techniques and issues related to the numerical simulation of free-surface flows. He points out that many tools exist which are adequate for dealing with the simpler cases in which the phases (be they fluid or solid) are coupled only through the exchange of momentum (see Hu *et al.* 2001, and Bunner & Tryggvason 2002, for example). The next step must be the development of techniques capable to describe more complex physics: heat transfer, phase change, electric and magnetic fields, and others.

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WORKSHOP ON SCIENTIFIC ISSUES
IN
MULTIPHASE FLOW

University of Illinois
at
Champaign-Urbana
May 7-9, 2002

APPENDIX 4

REPORT OF STUDY GROUP ON MICROPHYSICS

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APPENDIX 4

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Abstract

This Report outlines scientific issues that involve microscopic phenomena in multiphase flows. A common theme is the need to understand the coupling between molecular scale phenomena and macroscopic phenomena. Phenomena to be discussed include boiling nucleation, contact line motion, flow regimes in microchannels, breakup and coalescence of fluid particles and jets, atomization and sprays, and the effects of surface active molecules and drag reducing polymers.

1. Introduction

The coupling between molecular and macroscopic phenomena is an important feature of many multiphase flows. For example, it is well known that relatively small concentrations of surfactants can promote the breakup of bubbles or drops in turbulent flows. However, there is also experimental evidence that some surfactants have the opposite effect. These phenomena are important since, for example, they affect the size distribution of bubbles in chemical reactors such as fermenters. There is no quantitative model that can predict whether a given surfactant will increase or decrease the average bubble diameter and the effect of the concentration of the surfactant on the bubble.

The difficulty in developing such models is due to: (1) the complexity of the macroscopic flows; (2) the lack of reliable molecular scale models that can describe the behavior of complex molecules; and (3) the incomplete understanding of interfacial chemistry and physics that is relevant for multiphase flows. The macroscopic flows are typically turbulent and involve

extremely complex, time-dependent interfacial shapes. This behavior may be even more complicated in micro-channels in which the flow regimes may be qualitatively different than in larger channels. Even if one considers only the macroscopic length scales, large Reynolds number flows can easily span 3 or 4 orders of magnitude in eddy sizes.

In many situations, the motion of contact lines is important. For example, let us consider the motion of a drop on a smooth surface in the absence of phase change. One can describe the motion of the liquid inside most of the drop with the Navier-Stokes equation. However, there is a breakdown of continuum theory near the contact line. In this region, conventional theory would predict a singularity. Several researchers have argued that the no-slip boundary condition at the liquid-solid interface should be relaxed near the point of contact and replaced with a given slip boundary condition. This introduces an additional parameter, involving the ratio of a molecular length scale to the drop radius, into the mathematical description of the problem. However, recent experiments involving rapid condensation of steam onto surfaces with wettability gradients (Daniel *et al.* 2001) indicate that the motion of a drop can be dramatically different from its motion in the absence of condensation. To understand this phenomenon as well as others to be discussed later, significant improvements in the understanding of the effects of condensation and evaporation on the motion of contact lines are needed. A related issue is the phenomenon of contact line pinning by chemical impurities or roughness on solid surfaces.

Heterogeneous nucleate boiling on solid surfaces is another area in which a better understanding of surface chemistry and physics is needed. Conventional theories focus on the effects of small cavities on the nucleation of bubbles. However, when one considers nucleate boiling on extremely smooth surfaces, other effects may be important. Specifically, if one considers the boiling of water or other highly polar liquids on smooth surfaces, there is some evidence that small hydrophobic "islands" on the surface play an important role in nucleating bubbles. There are a number of intriguing observations of the effects of various treatments that require a better understanding before one can reliably predict the performance of smooth heater surfaces.

The expression “microphysics” may also be understood to include phenomena that, while strictly macroscopic, involve phenomena that are qualitatively different from those that are observed in traditional engineering systems. In the field of multiphase flows, microchannels provide an important illustration of this notion. In a microchannel, phenomena such as interfacial tension and wettability play a far more important role than in conventional channel flows. Associated with this are flow regimes such as “froth flow” that have no counterpart in larger channels. There are important issues, such as the influence of sharp corners and the effects of wettability of surfaces, that must be understood before one can take full advantage of microchannels in numerous heat transfer and other applications.

One tool that may be useful in addressing some of the above issues is the molecular dynamics (MD) simulation. Molecular dynamics (MD) simulations have improved with the advances in computational power since the early 1960's. Early models considered two-dimensional hard disks. Later improvements incorporated attractive forces and three-dimensionality. Recent advances have included sophisticated models of polar molecules, including water (da Rocha *et al.*. 2001). Such techniques are very promising for future work in several areas of multiphase flow science.

Several experimental tools have strong potential. These include particle image velocimetry (PIV), infrared thermometry, x-ray radiography, ultraviolet fluorescence, atomic

force microscopy (AFM), scanning electron microscopy (SEM), x-ray diffraction spectroscopy (XDS), and others that are discussed in the papers that are referenced in this Report.

In what follows, a set of critical scientific issues in this area of research will be elaborated.

2. Critical scientific issues

2.1 *Boiling nucleation*

Although much progress has been made in understanding the physics of forced convective boiling, there are many unresolved issues. There are two major problems related to the microphysics of two-phase flows: the modeling of forced convective subcooled nucleate boiling on the wall of the heated rods and the prediction of the bubble diameter (Delhaye 2002).

The nucleation of bubbles may occur either homogeneously or heterogeneously. In homogeneous nucleation, the bubbles appear spontaneously, without pre-existing interfaces, in the bulk of the liquid or on the wall. One would expect this to occur at the location of the spinodal curve, which is the locus of the minima of isotherms on the pressure-specific volume thermodynamic diagram. In most cases, the homogeneous nucleation temperature is approximately $0.9T_c$, where T_c is the critical temperature.

In practice, heterogeneous nucleation is more important than homogeneous nucleation because the superheats associated with heterogeneous nucleation are much smaller than those for homogeneous nucleation. Heterogeneous nucleation is generally associated with small cavities that contain trapped air or other non-condensable gases or vapor and they have sizes that are much larger than the critical radius for nucleation (Dhir 2002). Fig. 1 shows the nucleation of bubbles on an artificial cavity on the surface of a silicon wafer. The cavity has a square cross section with a side equal to 10 μm, and it is 100 μm deep. The cylinders in the picture are thermocouples; they are 1.58 mm in diameter.

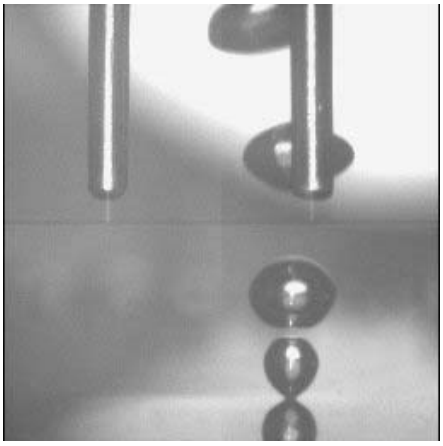

Fig. 1. Nucleation of vapor bubbles on an artificial cavity. (Courtesy of V. Dhir.)

The volume of gas trapped in a cavity depends on the magnitude of surface tension, the contact angle, the shape of the cavity, and the experimental conditions such as system pressure, liquid temperature, temperature of the heated surface, and dissolved gas content in the liquid.

The wall temperature at which nucleate boiling occurs depends strongly on the availability of cavities with trapped air (unflooded cavities.) Thus, as unflooded cavities become fewer and their size decreases, one might expect the heterogeneous nucleation temperature to approach the homogeneous nucleation temperature. To predict the number of active sites on a heated surface, one must know the number and size of cavities with trapped gas. By minimizing the Helmholtz free energy of a system involving the liquid-gas interface in the cavity, one can develop a criterion (Wang & Dhir 1993) that a cavity will trap gas if the contact angle, ϕ, is greater than the minimum cavity angle, ψ_{min}. For simple shapes, such as spherical or conical cavities, it is feasible to use this criterion. However, for a commercial surface, it would be necessary to have a knowledge of the size and shape of all cavities present on the surface in order to apply the above criterion.

It has been suggested (Mizukami 1975; Nishio 1985; Wang & Dhir 1993) that the instability of the gas/vapor nuclei in a cavity determines the incipience superheat. The vapor nucleus is stable if the curvature of the interface increases with an increase in vapor volume. Otherwise, the vapor bubble nucleus is unstable. Using this criterion, one may obtain an expression for the incipience superheat. The applicability of this criterion to copper surfaces of different wettabilities has been demonstrated (Wang & Dhir 1993) using water as the test liquid. The wettability was varied by oxidizing the surface.

The most difficult task is to predict the density of active nucleation sites. The active nucleation sites not only determine the phase structure adjacent to the heated surface, but also determine the mechanisms that contribute to the total heat transfer from the wall. The wall heat flux split between the liquid and vapor phases is also strongly influenced by the activity that takes place at a nucleation site. The following issues require further investigation:

- The physics of the process of entrapment in cavities of various sizes and shapes with advancing and receding interfaces, including the role of surface wettability.
- The effect of dissolved gases in the liquid on nucleation at the wall.
- The effect of absence of dissolved gases on the scavenging of trapped gas in the cavities.
- The behavior of the trapped gas-liquid interface during heating and up to the evolution of the embryo into a bubble.
- The activation/deactivation of nucleation sites due to temperature fluctuations in the solid and due to advancing and receding liquid-vapor interfaces over the cavities.
- The effect of various system variables such as pressure, liquid temperature, and flow velocity and properties of the solid on the above processes.

Another set of issues pertain to boiling nucleation on smooth surfaces. On such surfaces, the pre-existing nuclei (PEN) theory is not helpful. Experiments on nanoscopically smooth surfaces have been reported (Theofanous *et al.* 2002). On such surfaces, molecular scale surface inhomogeneities may be expected to play an important role in heterogeneous nucleation. Specifically, the BETA experiment (Dinh & Theofanous 2002) shows that nucleation of bubbles on titanium film heaters occurs at superheats equal to 5 to 7 K. The surfaces are 150 nm thick and have rms surface roughnesses equal to 4 nm. These observations are not consistent with the PEN theory and indicate the need to account for molecular scale phenomena. Further evidence for this idea is supplied by aging experiments in which the density of nucleation sites dramatically increases and one also observes an increase in the density of oxide islands on the surface. These experiments and others (Dinh & Theofanous 2002) provide motivation for another set of issues that require investigation:

- Based on the fact that heterogeneous nucleation occurs even on nanoscopically smooth surfaces, there is a need for new mechanistic approaches for treatment of heterogeneous nucleation in boiling on heater surfaces.
- Characterization of a heater surface nanomorphology and chemistry is necessary for a basic understanding of heterogeneous nucleation sites.
- Control of the experimental conditions and use of advanced instrumentation, such as infrared thermometry, x-ray radiography, and ultraviolet fluorescence, that enable accurate identification of nucleation events at high spatial and temporal resolution are needed.
- It is important to understand the dynamics of thin films (disjoining pressure, contact line motion) on a complex, non-uniform microscopic substrate. Such a substrate may be composed of either a Frenkel island (a small hydrophobic patch) or a Kuni nucleus (a site where vapor begins to form a drop.)
- In order to develop an understanding of boiling in micro-channels, it is critical to develop a basic understanding of nucleate boiling under geometrical constraints.
- It is of practical importance to determine whether nucleation in pool boiling is a self-limited response of a self-organized system to the externally imposed heat flux.
- How does hydrophobicity affect nucleation on nanoscopically smooth, cavity-free surfaces, and how can one use such knowledge to pattern a heater surface to control the coolability limit in boiling?
- How does exposure to air affect nucleation on nanoscopically smooth, cavity-free heater surfaces?

2.2 *Contact lines*

The importance of contact line motion in boiling nucleation has been mentioned in the previous subsection. The intermolecular interactions in the three phase contact line region are also important in many other situations such as adsorption, spreading, evaporation, condensation, wetting, and stability. Even if one considers spreading or sliding of drops on surfaces under isothermal conditions, there are issues that are not well understood at the present time. For example (Chen *et al.* 1997), measurements of contact angles at length scales of order 10^{-2} mm have been made by PIV, video microscopy, and digital image analysis. The results of these studies underscore the difficulty of characterizing or even defining the contact angle. Theoretical ideas about the static and dynamic contact angle and the role of intermolecular forces have been proposed (de Gennes 1985). However, at present, there are no direct experimental investigations of contact angles at the molecular scales.

A related issue is the phenomenon of contact line pinning. It has been speculated that pinning occurs when a moving contact line encounters a geometrical irregularity or a hydrophobic patch (de Gennes 1985). However, there are no direct experimental tests of these ideas or quantitative models that can predict when pinning will occur.

Condensation and evaporation introduce additional issues that are poorly understood at present. For example, a recent study (Daniel *et al.* 2001) considered the condensation of steam droplets onto a chemically treated silicon surface. The chemical treatment consisted of exposing the surface to silane vapor from a drop suspended a short distance above the surface. The silane molecules adsorbed onto the silicon and caused it to become hydrophobic. As a result, a contact angle gradient was created that caused small water drops to migrate toward more hydrophilic regions. Normally, the migration speeds of small drops is O(1 mm/s). However, when steam

condensed on the surface, the migration speeds were O(1 m/s). This dramatic increase in speed was, apparently, related to the condensation process; when the steam supply was shut off, the migration speeds of the drops on the surface quickly dropped to their normal values.

Although a few speculations have been made about the above phenomenon, there is no existing theory that can explain it. This points to the need to develop a deeper understanding of contact line motion that incorporates the effects of condensation and evaporation. Such understanding would find immediate application to heterogeneous nucleate boiling. A number of issues must be considered in developing theories for equilibrium and non-equilibrium situations (Wayner 2002). A detailed discussion may be found in several published papers (Potash and Wayner 1972; Reyes & Wayner 1996; Wayner 1991, 1994, 1999; Zheng *et al.* 2002). In general, improved modeling and experimental evaluation of the coupling of temperature and cohesion/adhesion on the transport processes in the contact line are needed. Insufficient emphasis has been placed on interfacial effects (both stress and temperature with polar and apolar systems) in very small systems and how they affect the macroscopically viewed transport processes. Innovative use of interfacial chemistry can lead to improved processes and devices.

In sum, the following issues should be pursued:

- A fundamental understanding of the significance the contact angle for both static and dynamic situations is needed.
- A quantitative theory of contact pinning that incorporates both surface chemistry and roughness should be developed.
- The relative importance (and coupling) or viscous stresses, slip, surface diffusion, molecular kinetics/dynamics, evaporation/condensation, excess free energy, contact angle, and film shape on contact line motion should be established.
- It is necessary to evaluate the adsorption isotherm for vapor/substrate systems of importance of phase change situations (e.g., boiling and rewetting of a hot surface) and connect these results with macroscopic phase change studies.
- For partially wetting systems, dropwise condensation should be studied and related to the apparent contact angle with heat flux, contact line motion, and the free energy of the thin film region.
- The importance/effect of surface chemistry relative to roughness on phase change processes should be established.
- It would be of great practical value to determine whether it is feasible to develop desirable and robust surfaces with surface chemistry.

2.3 *Microchannels*

Networks of micro-channels with hydraulic diameters in the 100 μm to 1 mm range have the capability for sustaining very large transport process rates in small volumes and are expected to play an important role in future energy systems such as fuel cells. Two distinguishing features of micro-channel flows in energy systems of interest are: (1) the importance of capillary effects relative to gravitational effects; and (2) the importance of fluid inertia (Ghiaasiaan 2002). One may characterize these effects by introducing the Eötvös (Eo), phase Weber (We_{LS}, and We_{GS}), and phase Reynolds numbers (Re_{LS} and Re_{GS}) for the channel, with the latter two parameters defined based on gas and liquid-phase superficial velocities. Detailed discussions of two phase flow in microchannels may be found in a recent paper (Ghiaasiaan & Abdel-Khalek 2001).

The parameter *Eo* compares the importance of gravity and surface tension. For the flows of interest, $Eo<1$, implying small buoyancy effect. However, as a consequence of the large flow velocities, the Reynolds and Weber numbers are both much larger than unity. Typical orders of magnitude are $Re>1$ and $We\sim10^2$. The significance of the large Weber number is that, unlike flow in porous media, inertia is important, and the liquid-gas interface can have complex topologies. The hydraulic diameter in these microchannels is typically smaller than the Laplace length scale, itself representing the order of magnitude of the interfacial wavelength associated with Taylor instability. The Taylor instability-driven interfacial phenomena, while crucial to many two-phase flow and change-of-phase processes in large channels, may thus be irrelevant to microchannels. Furthermore, velocity and temperature gradients in microchannels are much larger than those observed in large channels, leading to relative time and length scales, and relative magnitudes of interfacial forces that are different than large channels. Another difference between micro-channel flows and macroscopic flows lies in the relative importance of contact line motion in the former systems. This effect, while present in macroscopic flows, is likely to be much more important in micro-channels, in particular when heterogeneous nucleation and boiling phenomena are concerned.

Past experimental studies are limited in scope and detail. With respect to adiabatic two-phase flow, these studies indicate that all major and common two-phase flow regimes occur in microchannels. However, their details, and conditions leading to various regime transitions, are different than for large channels, and macroscale models and correlations often do poorly when compared to microchannel data. As an example, Fig. 2 shows plug-slug flow in a 1 mm tube.

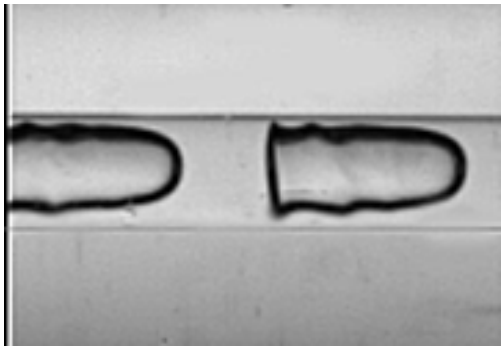

Fig. 2. Plug-slug flow in a 1 mm diameter tube flow. (Reprinted with permission from *Int. J. Multiphase Flow.*)

Furthermore, forced flow boiling processes appear to be significantly different than in large channels, primarily due to the potentially different heterogeneous bubble nucleation and boiling processes in microchannels (Ghiaasiaan 2002).

The critical issues pertaining to microchannels may be organized into two categories: issues for adiabatic flows and issues for flow boiling. For adiabatic flows, the issues are as follows:

- Experimental data covering a reasonable range of channel sizes (below 1 mm, in particular); and fluid pairs other than air/water-like pairs, are needed. The effects of liquid/gas density and viscosity ratios, surface tension, and surface wettability, in particular need to be examined.

- Channel geometries that contain multiple sharp corners are likely to be used extensively in practical applications. Data relevant to these flow passages are scarce and inadequate, however. Wettability of sharp corners may be particularly important.
- Flow regime identification has primarily been based on visual and photographic methods, and that has lead to much confusion. Objective methods for flow regime identification, to complement visual and photographic methods, or replace them when the test channel is not transparent, are needed.
- Flow regime details need to be investigated. This is particularly important for froth and annular flow patterns where disagreement between data and macro scale models is greatest. Details of annular flow are also important for understanding dryout in microchannels.

For flow boiling, the issues are as follows:

- Heterogeneous nucleation and boiling phenomena in microchannels subject to forced flow, and the behavior of micro bubbles after they are released, are unknown. Trends in the available forced flow boiling data suggest that these processes may have significant differences with what is known about macroscale boiling. Resolution of these issues is essential for understanding the apparent anomalies in the microchannel boiling curves, and other aspects of microchannel behavior.
- The details of the annular flow regime, and the phenomenology of processes that contribute to dryout need to be systematically investigated. Once again, trends in the available microchannel dryout data suggest a drastically different phenomenology than what is known about dryout in macroscale.

2.4 *Breakup and coalescence*

Breakup and coalescence of fluid objects play a crucial role in a broad spectrum of multiphase flow processes. Important examples include (Hanratty 2002; Longmire 2002; Reitz 2002):

- evolution of the bubble size distribution in stirred tanks and bubble columns
- drop size distribution in liquid emulsions
- breakup of liquid jets and the formation of sprays
- atomization of liquid layers in two phase flows.

The stumbling blocks to major breakthroughs in the above issues include microphysics issues as well as issues covered by the other Workshop Thrust Groups. Therefore, while this section will focus on issues that are primarily microscopic in nature, some overlap with the Reports for the other Thrust Groups is inevitable.

Perhaps the most relevant issues pertaining to breakup and coalescence of fluid objects are the microscopic conditions at the points of breakup and coalescence. Breakup of drops and bubbles involves intermolecular forces acting at micro or nanoscales. For example, if one considers a highly deformed bubble in a liquid, breakup may occur if a sufficiently thin liquid neck forms between two portions of the bubble. When the neck is sufficiently thin, intermolecular forces between the liquid molecules cause the neck to pinch off and two child bubbles result. In this respect, bubble breakup is similar to the coalescence of liquid drops in a gas. In the case of liquid breakup, intermolecular forces acting at micro and nanoscales must be overcome to separate neighboring layers of molecules. While the importance of molecular scale

phenomena is clear, there are no predictive criteria or models that can be used to obtain quantitative results.

The above issues are directly relevant to computer simulations of breakup and coalescence. Some computer simulations perform computational "surgery" to implement breakup or coalescence. Front tracking methods, which are relevant over a broad spectrum of flow Reynolds numbers (Nobari *et al.* 1996; Nobari & Tryggvason 1996), and boundary element methods, which are relevant for small Reynolds numbers, fall into this category. Other methods, such as VOF, level set methods, the phase field and second gradient methods, and the lattice Boltzmann method (LBM) allow breakup and coalescence to occur automatically. Fig. 3 shows a lattice Boltzmann simulation of bubble breakup in homogeneous turbulence (Qian *et al.* 2002).

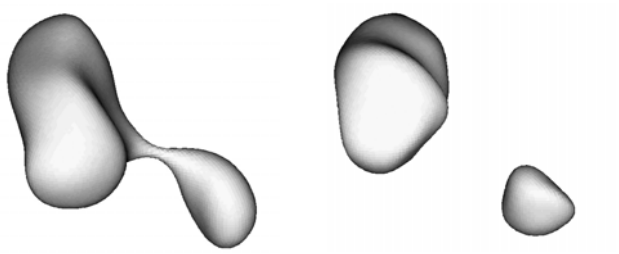

Fig. 3. A lattice Boltzmann simulation of bubble breakup in homogeneous turbulence.

With any of the above simulation methods, the critical issue is accuracy. The VOF and Level-Set methods handle coalescence and breakup "automatically" when the resolution of a thin film (or a thread) becomes less than a grid space or so. In this respect, they are probably similar to the Lattice Boltzmann method. This results in a rupture that depends strongly on the resolution, and it is therefore possible that low resolution results are qualitatively (not just quantitatively) different from those obtained with finer resolution. In the front tracking method one can explicitly specify a rupture criterion that does not directly relate to the resolution. These issues are discussed in more detail in a recent publication (Tryggvason *et al.* 2001). Clearly, it is extremely important that experimental and theoretical criteria for breakup and coalescence should be established.

Another issue that is directly related to the above topic is the role of surface active molecules on the above processes. It is well known that surfactants, even when present in relatively small concentrations, can strongly affect the coalescence or breakup of fluid objects. However, no predictive models are available. For example, consider the breakup and coalescence of bubbles in turbulent flows. This process has been studied (Walter & Blanch 1986) by measuring the size distribution of bubbles in a turbulent pipe flow. It was found that the effects of surfactants on the maximum stable bubble size were complex. One might expect that, by decreasing the surface tension, the maximum stable bubble size would decrease. However, in some cases, Walter and Blanch found the opposite effect. They were able to establish a rough criterion based on molecular weight. Surfactants of sufficiently small molecular weight always reduced the maximum stable bubble size, but surfactants with larger molecular weights increased the maximum stable bubble size when they were present in sufficiently small concentrations. It is not clear that one can generalize Walter and Blanch's findings to other surfactants. It is also not clear what causes the increase in the maximum stable bubble size in some cases. The increase

may be related to the Marangoni stresses found in simulations of bubble deformation in axisymmetric strain flows (Stone & Leal 1990). However, it must be pointed out that the simulations were restricted to Stokes flow, while the experiments involved bubble Reynolds numbers of $O(10^3)$. Also, the surfactant molecules were assumed to be insoluble in the liquid so that sorption kinetics were not considered in the simulations.

One can identify the following critical scientific issues pertaining to breakup and coalescence and related issues:

- Experiments and theoretical analyses are needed to establish rigorous criteria for the coalescence and breakup of fluid objects at the microscopic level. Without such criteria, current numerical simulation methods can, at best, be tested by indirect means.
- Experiments and molecular level simulations are needed to understand the role of surface active molecules on breakup and coalescence.
- In coalescence, the macroscopic instabilities and events leading to rupture in real flows are not well characterized. For example, the individual effects of inertia, shearing, interfacial tension, and three-dimensional flow perturbations on the coalescence process are not well understood, especially in geometries where many 'droplets' interact, such as dense sprays.
- It is important to develop theoretical frameworks that will enable one to couple macroscopic and microscopic behaviors into a physically meaningful and accurate model.
- For engine applications, the correlations used in collision and coalescence models need to be tested against fundamental experiments at high pressures since they come from sprays in entirely different regimes (rain drops and cloud physics).
- The relationship between drop breakup and drop shattering, and turbulence production and dissipation in sprays needs to be established.
- More experimental data is needed in order to improve wall impingement models. In particular, data on the secondary breakup of impinged drops is needed under high wall temperature (T_w>400 K), gas temperature (T_g>700 K) and density (ρ>20 kg/m^3) conditions. More accurate models for the thickness of the wall film also need to be developed. Improved models should be able to model the spreading of a liquid film, and should account for the effect of wall wetting and liquid splashing.

2.5 *Effects of surfactants on interfacial flows*

In the previous section, it was pointed out that surfactants have a significant effect on breakup and coalescence phenomena. It is also well known that they strongly affect the rise velocity of bubbles and drops (McLaughlin 2002). For example, concentrations as low as 10^{-5} mol/m^3 of common surfactants can reduce the rise velocity of a 1 mm bubble by more than 50% (Zhang *et al.* 2001). This effect has been attributed to the formation of an immobilized surfactant cap on the bubble surface (Levich 1962; McLaughlin 1996). Such changes not only affect bubble holdups in chemical reactors, but also dramatically reduce mass transfer rates (Lochiel & Calderbank 1964). The observation of a significant enhancement of heat transfer in the pool boiling of surfactant solutions is also noteworthy (Hetsroni *et al.* 2001; Hetsroni & Mosyak 1999). Clearly, these behaviors call into question the usefulness of simulations or models that do not consider the influence of surface-active materials on the behavior of systems with air-water interfaces.

Although it is well known that low concentrations of surfactants can have dramatic effects on simple two-phase flows, relatively little is known about their effects on complex two-phase flows. For example, there are numerous experimental results for the effects of surfactants on freely rising bubbles or on the damping of capillary waves, but relatively little is known about the effectiveness of surfactants in turbulent two-phase flows. A better understanding of the conformation, sorption kinetics, and effects of surfactants on interfaces would impact a large range of energy-related technologies including bioreactors, flotation and microflotation, enhanced oil recovery, sedimentation and filtration, and cleaning processes. The effects of surfactants on surface waves may also play a role in the sequestration of atmospheric carbon dioxide in the oceans because of their effect on capillary waves.

Many investigators (Chang & Franses 1995) have modeled the sorption kinetics of surfactants onto interfaces with a two parameter model suggested by Langmuir. The parameters are the adsorption rate constant, β (m^3/mol-s), and the desorption rate constant, α (s^{-1}). The ratio of these constants may be determined by equilibrium measurements of the surface tension as a function of the concentration of surfactant. The actual values of the parameters are determined by dynamic surface tension measurements. It is known that the parameters depend on the surfactant concentration in the liquid, C (mol/m^3), as well as the surfactant concentration on the interface, Γ (mol/m^2). To model the effects of the adsorbed surfactant on the surface tension, γ, an equation of state is needed. For very small values of Γ, an ideal gas equation of state (e.g., Stone and Leal 1990) is used. At larger concentrations, an equation of state (Frumkin 1925) that involves a parameter, Γ_m, that represents the maximum possible surface concentration.

There are several issues that must be addressed in order to use the above modeling to describe moderate or high Reynolds number two phase flows. First, the sorption rate constants are available only for a relatively small number of surfactants. Secondly, the sorption rate constants are known only in relatively small concentration ranges. These concentration ranges are dictated by the fact that, to obtain values of the sorption rate constants, one must perform experiments for conditions in which adsorption on the gas-liquid interface is the rate limiting step. In the past, such experiments were typically carried out at small Reynolds numbers and at fairly large concentrations of surfactant so that the liquid near the gas-liquid interface did not become depleted (which would make boundary layer mass transfer the rate limiting step.) However, in many situations of interest, the Reynolds numbers are large and the concentrations of surfactant are much smaller than the concentrations for which experimental results for the sorption rate constants are available.

Recently, a possible solution to this problem has been suggested (Liao & McLaughlin 2000; Wang 2001; Wang *et al.* 2002). The idea is to measure the time-dependent rise velocity of a bubble in low concentration surfactant solutions and to compare the results with simulations that account for surfactant transport in the liquid and on the interface as well as sorption rate kinetics (Liao & McLaughlin 2000). Measurements of bubble rise velocity for single bubbles in very dilute aqueous solutions of the non-ionic surfactant Triton X-100 show that the bubble achieves a maximum velocity that is close to the terminal velocity in pure water (Duineveld 1995) before slowing down to its terminal velocity. Eventually, the rate of surfactant adsorption on the bubble surface becomes mass transfer limited (Zhang *et al.* 2001). However, at earlier times, sorption rate constants have a strong effect on the motion of the bubble. For example, during this initial period, bubbles in SDS solutions slow down an order of magnitude more gradually than in Triton X-100 solutions at the same molar concentration (Wang 2001; Wang *et al.* 2002). Thus, by focusing on the first few seconds of the bubble's motion, one can observe

differences attributable to the sorption kinetics. Preliminary results are available. For example, values for the sorption rate constants have been obtained for Triton X-100; previously, only lower bounds (at a much larger surfactant concentration) were available. It appears feasible to obtain results for other surfactants using the same approach.

A further complication is the effect of unsteadiness in the liquid phase. In the experiments discussed above, the only unsteadiness was that caused by the acceleration of the bubble. However, in situations of practical interest, the surrounding flow is likely to be turbulent as a result of stirring or processes involving other bubbles. This presents an additional challenge for modeling.

In sum, there are a number of scientific issues involving surfactants that should be addressed in order to make substantial progress in the understanding and modeling of two-phase flows:

- There is a need to obtain results for sorption kinetics rate parameters for surfactants for a wide range of concentrations.
- The usefulness of existing models of sorption kinetics in unsteady, high Reynolds number flows should be evaluated.
- The effects of surfactants on coalescence and breakup of bubbles or drops in turbulent two phase flows involving bubbles or drops are only crudely understood. Careful experiments that can identify the various regimes of behavior are needed. For example, experiments with pairs of bubbles in motionless liquids suggest that, for "small" concentrations of surfactants, coalescence is inhibited by the adsorbed surfactants.
- The behavior of adsorbed surfactant molecules on the surfaces of bubbles in turbulent flows is not known. The surfactant cap model works well for bubbles or drops translating through motionless fluids, but it is not clear that such a model is useful for bubbles in turbulent flow. Do adsorbed surfactant molecules inhibit mass transfer in such situations?

2.6 *Drag reducing polymers*

It is now established that the large-scale behavior of multiphase flows is dependent of small scale phenomena (Hanratty 2002). As our understanding of the microphysics improves, we have the opportunity to carry out basic studies in which methods for changing the behavior of multiphase flows are explored. One example is the use of structured tubes in heat transfer applications. However, most striking are recent experiments in which drag-reducing polymers were added to a gas-liquid flow.

These polymers were injected into a gas-liquid annular flow in a horizontal pipe. With mixture concentrations as low as 15 ppm, the violent annular flow could be changed into a stratified pattern with a relatively smooth interface and no entrainment. The explanation for this behavior is that the polymers destroy the disturbance waves. These waves are the loci at which atomization occurs; they also play an important role in causing the liquid to move up the walls of a horizontal pipe against the pull of gravity. More recent studies in Hanratty's laboratory have explored the influence of drag-reducing polymers on a highly agitated stratified flow for which the waves were so large that the interfacial drag was almost 20 times what would be observed for an interface without waves. The interfacial drag was greatly reduced, the liquid holdup was increased and, for increasing liquid flow rate, the transition to slug flow was delayed.

Experiments, such as these, show in a spectacular way that the macroscopic behavior of multiphase systems depends on small scale phenomena. They provide a test for our physical understanding and suggest new technological approaches.

A considerable literature exists on the influence of polymer additives on single phase flows. The number of investigations of their effect on gas-liquid flows is small (Rosehart *et al.* 1972; Otten & Fayed 1976; Kang *et al.* 1997, Sylvester & Brill 1976; Al Sarkhi & Hanratty 2001a, 2001b). Decreases in the pressure drops are noted, but the most interesting aspect of these studies is the finding that flow patterns and wave patterns can be dramatically changed. A recent paper (Manfield *et al.* 1999) reviews the subject.

Polymers are found to destroy the turbulence in slugs. One of the consequences of this is that larger liquid flows are needed to initiate slugging. Slugs pick up liquid at their front and shed liquid at the back. If the shedding rate is greater than the pickup at the front, the slug becomes unstable. The increase in the critical liquid rate to generate slugs can be interpreted by an increase in the shedding rate in a reference frame moving with the velocity of the fluid in the slug, u_s. The back of a slug is pictured as the nose of a large bubble. The velocity of this bubble relative to the velocity of the fluid in the slug, V_B/u_s, determines the shedding rate. A number of investigators have suggested that V_B/u_s equals the ratio of the centerline velocity of the fluid in the slug to the average velocity (Woods & Hanratty 1996). This gives $V_B/u_s \cong 1.2$ for a turbulent flow, in agreement with measurements of V_B. If the flow in the slug is laminar one could infer that $V_B/u_s \cong 1.5$. The change in V_B/u_s agrees with observations of an increase in the critical liquid height needed to generate slugs when polymers are added.

Another effect of polymers is the damping of waves. This gives rise to the destruction of disturbance waves. This, in turn, causes a change from an annular to a stratified flow, because atomization of the liquid layer ceases and because the ability of the liquid to climb up the wall is diminished. Polymers have also been observed to decrease the interfacial drag at the interface by changing wave properties. This, in turn, contributes to an increase in the liquid holdup.

The role of polymers in damping waves is not understood. One possibility is that elongated polymers at the interface introduce tensile stresses close to the interface and, therefore, have the same affect as increasing surface tension.

3. Conclusion

The issues summarized in this Report are significant scientific challenges. A common theme is the need to bridge the gap between the nanoscale, which can be studied with molecular dynamics simulations, and the micrometer scale, which in many cases, sets the limit on currently available experimental techniques, such as PIV, that can elucidate macroscopic fluid motion and phase distributions. The following recommendations emerged from the Workshop:

- An approach that requires equal contributions of experimentation, simulation, and modeling is needed to fill the gap (modeling) between nanoscale (simulation) and microscale (experimentation).
- The microphysics area is perhaps the most challenging area of the four focus groups because the tools for bridging the gap from nanoscale to microscale are the least developed.
- Collaborative research involving academia, the DOE laboratories, and industry should be strongly encouraged because of the complexity and scope of problem.

- Sharing of computer simulation programs and experimental databases should be encouraged.

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