A COMPARISON OF A KINETIC THEORY CFD MODEL FOR GAS-LIQUID-SOLID FLUIDIZATION TO EXPERIMENTS WITH GLASS BEADS AND METHANOL CATALYST

by

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ABSTRACT

The objective of this study is to develop a predictive experimentally verified computational fluid dynamic (CFD) three phase model. It predicts the gas, liquid and solid hold-ups (volume fractions) and flow patterns in the industrially important bubble-coalesced (churn-turbulent) regime. The input into the model can be either particulate viscosities as measured with a Brookfield viscometer or effective restitution coefficient for particles.

A combination of x-ray and γ-ray densitometers was used to measure solid and liquid volume fractions. There is a fair agreement between the theory and the experiment. A CCD camera was used to measure instantaneous particle velocities. There is a good agreement between the computed time average velocities and the measurements. There is an excellent agreement between the viscosity of 800 μm glass beads obtained from measurement of granular temperature (random kinetic energy of particles) and the measurement using a Brookfield viscometer.

A relation between particle Reynolds stresses and granular temperature was found for developed flow. Such measurement and computations gave a restitution coefficient for a methanol catalyst to be about 0.9.

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INTRODUCTION

Fluidized beds are widely used industrially because the particles can be introduced into and out of a reactor as a fluid and because of good heat and mass transfer in the reactor. The invention of kinetic theory of granular flow by professor Savage and collaborators (Savage & Jeffrey, 1981; Savage, 1983 & 1988; Lun et al, 1984), as reviewed by Gidaspow (1994), provides a plausible explanation for this fluid-like behavior of particles. The particles oscillate randomly. This assumption, supported by measurements (Carlos and Richardson, 1968; Gidaspow and Huilin, 1996 & 1997; Arastoopour and Yang, 1992; Cody, et al, 1996, 1997) allows the definition of granular temperature as one third the sum of the squares in the three directions of the random oscillations of particle velocities. In other words, the granular temperature is the random kinetic energy of particles per unit mass. It is directly related to the turbulent kinetic energy used in single phase flow in the k-epsilon model (Mohammadi and Pironneau, 1994) and extended to multiphase flow by Ahmadi and Cao (1990) and to gas-particle computation flow by Cao and Ahmadi (1995). The distinction is that in the granular flow theory this temperature acts like the thermal temperature in kinetic theory of dense gases and in statistical mechanics of fluids. This analogy allows an easy application of these theories to fluidized beds. For example, the kinematic viscosity of the fluidized beds can be computed from a direct measurement of the particle oscillations, as essentially the product of the mean free path and the oscillating particle velocity. This paper illustrates the application of this theory to gas-liquid-solid fluidized beds. Such computations for gas-solid flow in a riser were performed by Samuelsberg and Hjertager (1996).

Fan (1989) has reviewed three-phase fluidization and its applications. Tarmy and Coulaloglu (1992) show that there exist no hydrodynamic models for three phase fluidization in the literature. Reactor design using slurry bubble columns is usually done using hold-up correlations (Viking Systems International, 1994). Papers presented at the recent Computational Fluid Dynamics (CFD) in Reaction Engineering Conference held in San Diego in October 1996 by the EXXON (Heard, 1996) and the Los-Alamos groups (VanderHeyden , 1996) have described their work in using CFD models to understand the injection of oil into fluidized catalytic cracking risers. The application of interest in this study is the slurry bubble column reactor for making of methanol from synthesis gas. EXXON and other companies are commercializing processes to convert the apparently abundant natural gas into storable liquid fuels. Fluidized beds are often used in such applications. The CFD approach described in this paper should eventually help in the design of more efficient reactors, such as the selection of internals for proper flow patterns and bubble control and optimization of size of catalyst. For such a purpose an experimentally verified CFD model is needed.

PART I : HYDRODYNAMIC MODEL

The physical principles used are the laws of conservation of mass and momentum for the gas, the liquid and the solid phases, as shown in Table 1. The constitutive equations for the stress are also shown in Table 1. This approach is similar to that of Soo (1967) for multiphase flow and of Jackson (1985) for fluidization. The equations are similar to

Bowen's (1976) balance laws for multi-component mixtures. The principle difference is the appearance of the volume fraction of phase "k" denoted by ε_k. In the case of phases not all the space is occupied at the same time by all the phases, as it is by components. As in the case of the mixture equations for components, the mixture equations for phases show that the sum of the drag forces is zero. The fluid pressure p_l is in the liquid (continuous) phase.

The corresponding gas and solids (dispersed particulate phases) momentum equations with zero particulate viscosities are the trajectory equations for particulate flow. For gas-solid fluidized beds, Bouillard, et al. (1989) have shown that this set of equations produces essentially the same numerical answers for fluidization as did the earlier conditionally stable model which has the fluid pressure in both the gas and the solids phases. In this model (hydrodynamic model B) the drag and the stress relations were altered to satisfy Archimedes' buoyancy principle and Darcy's law, as illustrated by Jayaswal (1991). The gas-particulate drag coefficients given in Table 1 are for the model B reviewed by Jayaswal (1991). Note that Equation (2a) does not contain the volume fraction in the gravity term, while Equation (2b) has a buoyancy term. This is a generalization of model B for gas-solid systems as discussed by Gidaspow (1994) in section 2.4. For the solid phase, p_s consists of the static normal stress and the dynamic stress, called solids pressure, which arises due to the collision of the particles. The added mass forces and the Saffman's lift forces (Saffman, 1965; Drew, et al., 1979) were assumed to be negligible.

This model is unconditionally well-posed, i.e., the characteristics are real and distinct for one dimensional transient flow. It does not require the presence of solids pressure for stability and well posedness. The details of hydrodynamic equations used in the computer simulation are given in Table 1.

NUMERICAL SCHEME

The governing equations along with the constitutive equations are solved for p_l, ε_k, u_k and v_k (k = g, l, s) using the Implicit Continuous Eulerian (ICE) method (Rivard and Torrey, 1977, Jayaswal, 1991) with appropriate initial and boundary conditions. Stewart and Wendroff (1984) have critically reviewed the ICE algorithm and related staggered mesh conservative schemes. The computations are carried out using a mesh of finite-difference cells fixed in two-dimensional space (Eulerian mesh). The scalar variables are located at the cell center and the vector variables at the cell boundaries. The momentum equation is solved using staggered mesh, where as for continuity equation a donor cell method was used.

The partial differential equations are well-posed. The time step is chosen to satisfy the Courant stability criterion (Courant, et al., 1952). The numerical stability of the equations can be obtained using the von Neumann stability analysis, as illustrated by Lyczkowski, et al. (1978) and Prosperetti (1982).

TABLE 1. Governing Equations - Hydrodynamic Model B for Gas-Liquid-Solid Flows

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In the following equations, the tensor is represented as [ ].

1. Continuity Equation for Phase k *(= g, l, s).*

$$\frac{\partial}{\partial t}(\epsilon_k \rho_k) + \nabla\cdot(\epsilon_k \rho_k \boldsymbol{v}_k) = 0$$

2. Momentum Equations.

(a) Liquid Phase (Continuous Phase)

$$\frac{\partial}{\partial t}(\epsilon_l \rho_l \boldsymbol{v}_l) + \nabla\cdot(\epsilon_l \rho_l \boldsymbol{v}_l \boldsymbol{v}_l) = -\nabla p_l + \rho_l \boldsymbol{g} + \sum_{m=g,s} \beta_{lm}(\boldsymbol{v}_m - \boldsymbol{v}_l) + \nabla\cdot[\tau_l]$$

(b) Gas / Solid Phases k *(= g, s)* (Dispersed Phases)

$$\frac{\partial}{\partial t}(\epsilon_k \rho_k \boldsymbol{v}_k) + \nabla\cdot(\epsilon_k \rho_k \boldsymbol{v}_k \boldsymbol{v}_k) = -\nabla p_k + \frac{\epsilon_k}{\epsilon_l}(\rho_k - \sum_{m=l,g,s\ m\neq k} \epsilon_m \rho_m)\boldsymbol{g} + \sum_{m=l,g,s\ m\neq k} \beta_{km}(\boldsymbol{v}_m - \boldsymbol{v}_k) + \nabla\cdot[\tau_k]$$

3. Constitutive Equations for Stress.

a) Fluid Phase Stress. k *(= g, l)*

$$[\tau_k] = \epsilon_k \mu_k \left([\nabla \boldsymbol{v}_k + (\nabla \boldsymbol{v}_k)^T] - \frac{2}{3}\nabla\cdot(\boldsymbol{v}_k)\,[I] \right)$$

a) Solid Phase Stress.

$$[\tau_s] = \mu_s [\nabla \boldsymbol{v}_s + (\nabla \boldsymbol{v}_s)^T] + (\xi_s - \frac{2}{3}\mu_s)\nabla\cdot(\boldsymbol{v}_s)\,[I]$$

4A. Kinetic Theory Model.

Fluctuating Energy Θ_s (= ⅓ $< C^2 >$) Equation,

$$\frac{3}{2}\left(\frac{\partial}{\partial t}(\epsilon_s \rho_s \Theta_s) + \nabla\cdot(\epsilon_s \rho_s \Theta_s \boldsymbol{v}_s)\right) = -p_s \nabla\cdot\boldsymbol{v}_s + \Phi_k - \nabla\cdot\boldsymbol{q}_s - \gamma_s - 3\beta_{ls}\Theta_s$$

Collision Energy Dissipation Rate,

$$\gamma_s = 3(1-e^2)\epsilon_s^2 \rho_s d_s g_0 \Theta_s \left(\frac{4}{d_k}\left(\frac{\Theta_s}{\pi}\right)^{\frac{1}{2}} - \nabla \cdot \vec{v}_s \right)$$

Flux of Fluctuating Energy,

$$\vec{q}_s = -\kappa_s \nabla \Theta_s$$

Conductivity of Fluctuating Energy,

$$\kappa_s = \frac{2\kappa_{dil}}{(1+e)g_0}\left[1+\frac{6}{5}(1+e)g_0\epsilon_s\right]^2 + 2\epsilon_s^2\rho_s d_s (1+e) g_0 \left(\frac{\Theta_s}{\pi}\right)^{\frac{1}{2}}$$

Dilute Phase ("Eddy Type") Granular Conductivity,

$$\kappa_{dil} = \frac{25\sqrt{\pi}}{128}\rho_s d_s \Theta_s^{\frac{1}{2}}$$

Solid Phase Shear Viscosity,

$$\mu_s = \frac{2\mu_{dil}}{(1+e)g_0}\left[1+\frac{4}{5}(1+e)g_0\epsilon_s\right]^2 + \frac{4}{5}\epsilon_s^2\rho_s d_s (1+e) g_0 \left(\frac{\Theta_s}{\pi}\right)^{\frac{1}{2}}$$

Solid Phase Bulk Viscosity,

$$\xi_s = \frac{4}{3}\epsilon_s^2\rho_s d_s (1+e) g_0 \left(\frac{\Theta_s}{\pi}\right)^{\frac{1}{2}}$$

Dilute Phase Solids Viscosity,

$$\mu_{dil} = \frac{5\sqrt{\pi}}{96}\rho_s d_s \Theta_s^{\frac{1}{2}}$$

Solid Phase Pressure

$$p_s = \epsilon_s \rho_s \Theta_s [1 + 2(1+e)\epsilon_s g_0]$$

Radial Distribution Function,

$$g_0 = \left[1 - \left(\frac{\epsilon_s}{\epsilon_{s,\max}}\right)^{\frac{1}{3}}\right]^{-1}$$

Energy Dissipation Rate

$$\Phi_s = [\tau_s] : \nabla \mathbf{v}_s$$

4B. Empirical Solids Viscosity and Stress Model.

$$\nabla p_k = G(\epsilon_k)\, \nabla \epsilon_k$$

$$\xi_s = 0$$

$$\mu_s = 5\epsilon_s \ (poises) \qquad (example)$$

5. Continuous Phase-Particulate Phase Drag Coefficients. $k\ (= g, s)$
for $\epsilon_l < 0.8$, (based on Ergun equation)

$$\beta_{lk} = \beta_{kl} = 150\frac{(1-\epsilon_l)\epsilon_k \mu_l}{(\epsilon_l d_k \psi_k)^2} + 1.75\frac{\rho_l \epsilon_k |\mathbf{v}_l - \mathbf{v}_k|}{\epsilon_l d_k \psi_k}$$

for $\epsilon_l \geq 0.8$, (based on empirical correlation)

$$\beta_{lk} = \beta_{kl} = \frac{3}{4} C_D \frac{\epsilon_k \rho_l |\mathbf{v}_l - \mathbf{v}_k|}{d_k \psi_k} \epsilon_l^{-2.65}$$

where,

$$C_D = \begin{cases} \dfrac{24}{Re_k}(1 + 0.15 Re_k^{0.687}) & for\ Re_k < 1000 \\ 0.44 & for\ Re_k \geq 1000 \end{cases}$$

$$Re_k = \frac{\epsilon_l \rho_l |\mathbf{v}_l - \mathbf{v}_k| d_k \psi_k}{\mu_l}$$

6. Particle-Particle Drag Coefficients. *k, m (= g, l, s)*

$$\beta_{km\ k+m}=\frac{\alpha(1+e)\epsilon_k\rho_k\epsilon_m\rho_m(d_k+d_m)^2\left[1+3\left(\frac{\epsilon_{km}}{\epsilon_k+\epsilon_m}\right)^{\frac{1}{3}}\right]}{2\epsilon_f(\rho_k d_k^3+\rho_m d_m^3)\left[\left(\frac{\epsilon_{km}}{\epsilon_k+\epsilon_m}\right)^{\frac{1}{3}}-1\right]}\,|\,v_k-v_m\,|$$

where

$$\epsilon_{km}=\begin{cases}[(\phi_k-\phi_m)+(1-a)(1-\phi_k)\phi_m]\,[\phi_k+(1-\phi_m)\phi_k]\,\dfrac{X_k}{\phi_k}+\phi_m & \text{for } X_k\le X_{k,o}\\ (1-a)\,[\phi_k+(1-\phi_k)\phi_m]\,(1-X_k)+\phi_k & \text{for } X_k\ge X_{k,o}\end{cases}$$

$$a=\sqrt{\frac{d_m}{d_k}}\qquad (d_k\ge d_m)$$

$$X_k=\frac{\epsilon_k}{\epsilon_k+\epsilon_m}$$

$$X_{k,o}=\frac{\phi_k}{[\phi_k+(1-\phi_k)\phi_m]}$$

7. Definitions.

$$\epsilon_g+\epsilon_l+\epsilon_s=1$$

The liquid, gas, and solid phases are assumed to be incompressible and constant densities are used. The constitutive relations in the kinetic theory model were derived by Gidaspow (1994). The constitutive relations may be easily changed by some program modification.

DISPERSED BUBBLE FLUIDIZATION REGIME

Carlos and Richardson (1968) studied the movement of large particles in the liquid-solid fluidized bed and reported the measurements of the velocity components of particles. They used a cylindrical fluidization column made of standard 10.16 cm diameter Q.V.F. glass 121.92 cm in length and carried out the experimental work with uniform sized glass bead particles with dimethyl phthalate fluid. The diameter of glass bead particles used was 0.889 cm. The densities of glass bead particles and dimethyl phthalate fluid were 2.49 g/cm^3 and 1. 19 g/cm^3, respectively. Liquid was fed at the bottom of the bed. The particles in the bed were filled to a constant bed height of 26 cm. Initial superficial liquid velocity was 4.8 cm/s, which was the minimum fluidization velocity of the 0.889 cm glass beads in dimethyl phthalate.

In the three phase fluidization study, we used the above described fluidization system with gas injection at the bottom of the bed. The gas phase was treated as a particulate phase having 0.4 cm diameter dispersed spherical air bubbles at room temperature (27°C) with constant density.

Hydrodynamic model B with the kinetic theory model was used to simulate this gas-liquid-solid fluidized bed. The governing equations are shown in Table 1. The geometry and dimensions are given in Figure 1.

Uniform finite difference grids were used in computations. It was assumed that angular symmetry existed. The fluidized bed was divided into 154 computational cells. Each cell was 1.016 cm by 2.5 cm size. Numerical computations for fluidization were carried out for 1.5 seconds with a time increment of 5×10^{-6} seconds. The computations were conducted on Cray YMP/4-464 supercomputer at the National Center for Supercomputing Applications (NCSA) at University of Illinois, Urbana-Champaign, Illinois.

Four different cases for gas-liquid-solid fluidization system were simulated in this study. Flow conditions and the volume fractions used in each simulation case are given below in Table 2.

TABLE 2. Flow Conditions for Dispersed Bubble Flow Regime

U_{mf}=4.8 cm/s; height of fluidized bed=26 cm

Cases	$U_{sup,l}$ (cm/s)	U/U_{mf}	Bed Volume Fraction	Inlet Gas Volume Fraction	$U_{sup,g}$ (cm/s)
1	7.6	1.6	0.53	0.03	0.844
2	9.4	2.0	0.58	0.03	0.844
3	9.4	2.0	0.58	0.07	1.968
4	15.1	3.1	0.70	0.07	1.968

The particle terminal velocity ($v_{s,t}$) for glass beads was 53.7 cm/s and gas bubbles terminal velocity ($v_{g,t}$) was 28.1 cm/s.

Results For Dispersed Bubble Regime.

In this regime, no experimental work was performed in our laboratory. A transient three phase fluidized behavior inside the bed was simulated. The phase holdup in the bed for three phase fluidization was computed and compared with the estimated results from a number of existing empirical correlations for phase holdups given by Saberian-Broudjenni, et al. (1984); Sinha, et al. (1984); and Dakshinamurthy, et al. (1972). The correlations are not repeated here as they are summarized in Table 2.1 by Fan (1989). Good comparison was obtained as shown in Table 3.

TABLE 3. Average Gas-Liquid Volume Fraction in Bed

Cases	Computed	Saberian-Broudjenni, et al. (1984)	Sinha, et al. (1984)	Dakshinamurthy, et al (1972)
1	0.572	0.580	0.576	0.486
2	0.623	0.615	0.602	0.552
3	0.639	0.647	0.633	0.591
4	0.752	0.735	0.695	0.785

No bubble coalescence was observed in the simulated three phase fluidization. The gas phase remained dispersed in all four cases. This result is consistent with the predictions from flow regime diagram for concurrent gas-liquid-solid fluidized bed shown in Figure 2.3 (Fan, 1989) for bed diameter of 10.16 cm, and particles terminal velocity of 53.7 cm/s, although care must be taken because Figure 2.3 (Fan, 1989) was prepared for air-water-particle fluidization system.

Figure 2 shows typical computed granular temperature and shear viscosity inside the bed. In general, the local solids shear viscosity varies between 1 and 20 poises for all the simulation cases. The shear viscosity is higher at the lower section of the bed and close to the wall and decreases to zero in the top section of the bed. In the top section the shear viscosity is almost constant across the cross-section of the bed. The granular temperature inside the bed also shows considerable fluctuations in the lower section of the fluidized bed.

Earlier, for the gas-solid systems, solid shear viscosity was computed from experimental measurements to be of the order of $5\varepsilon_s$ poises (Gidaspow, 1986). Song and Fan (1986) measured apparent viscosity for air-water-glass beads system and obtained values ranging from 0.5 to 15 poises. The apparent viscosity correlation for three phase fluidized bed developed by Song and Fan (1986) gives values ranging from 15 to 20 poises. These values compare favorably with the solids viscosity shown in Figure 2.

The computed velocity patterns for each of the four simulation runs are similar and only the velocity patterns for Case 1 (see Table 2) is shown in Figure 3. There is considerable scatter in computed velocities. A general velocity pattern within the bed is upward movement of particles in the center and downward movement at the walls. There is marked inward radial movement near the bottom of the bed and outward radial movement higher in the bed.

In the dispersed bubble three phase fluidization regime, no comparison with the experiment is available. However, in the same fluidization system with no gas flow, i.e., for liquid-solid fluidization, fair comparison between experimentally measured (Carlos and Richardson, 1968) and computed solid velocities (both axial and radial components) were obtained, as reported elsewhere (Gidaspow, et al., 1991).

PART II: COMPARISON TO EXPERIMENT WITH GLASS BEADS IN BUBBLY COALESCED FLUIDIZATION REGIME

Experimental Setup.

The setup used in the bubbly coalesced regime for volume fraction, velocity and viscosity measurement experiments consisted of four major parts: fluidization equipment, densitometers assembly, a high resolution micro-imaging / measuring system or a video-digital camera unit, and a Brookfield viscometer. A schematic diagram of the fluidized bed and video-digital camera unit for velocity measurements is shown in Figure 4. The schematic diagram for source-detector-recorder assembly for x-ray and γ-ray densitometers for volume fraction measurements is shown schematically in Figure 5.

a. Fluidization Equipment. A two-dimensional bed was constructed from transparent acrylic (Plexiglas) sheets to facilitate visual observation and video recording of the bed operations such as gas bubbling and coalescence, and the mixing and segregation of solids. The bed height was 213.36 cm and cross-section was 30.48 cm by 5.08 cm. A centrifugal pump was connected to the bottom of the bed by a 1.0 inch (2.54 cm) diameter stainless steel pipe. Gas injection nozzles from an air compressor were connected to the sides of the bed. Liquid was stored in and recycled back to a fifty five gallon storage tank.

The liquid and gas distributors were located at the bottom of the bed. The liquid was distributed by two perforated Plexiglas plates with many 0.28 cm diameter holes. They were placed at 35.6 cm and 50.8 cm above the bottom of the bed, with 0.25 cm size glass bead particles filled inside. The gas distributor consisted of six staggered porous tubes of 15.24 cm length and 0.28 cm diameter. The fine pores of porous tubes had mean diameter of 42 μm. The porous tubes were placed at the bottom of the bed just below the top liquid distributor plate.

b. Densitometer Assembly. Two densitometers were used alternatively for measuring the time-averaged volume fractions of three phases at a designated location by means of the x-ray and γ-ray adsorption techniques. The assembly consisted of radioactive sources

as well as detecting and recording devices and a positioning table. A schematic diagram of the source, detector and recording devices assembly is shown in Figure 5.

(1) Radioactive Source. The source is a 200-mCi Cu-244 source having 17.8-year half-life. It emitted x-rays with a photon energy between 12 and 23 keV. The source was contained in a ceramic enamel, recessed into a stainless steel support with a tungsten alloy packing, and sealed in welded Monel Capsule. The device had brazed Beryllium window. For the γ-ray densitometer, a 20-mCi Cs-137 source having a single γ-ray of 667 keV and a half-life of 30 years was used. The source was sealed in a welded, stainless steel capsule. The source holder was welded, filled with lead, and provided with a shutter to turn off the source. This is the same unit used previously by Gidaspow, et al. (1983).

(2) Detecting and Recording Devices. The intensity of the x-ray beam was measured by using a NaI crystal scintillation detector (Teledyne, ST-82-1/B). It consisted of a 2-mm thick, 5.08 cm diameter tube with 0.13-mm thick Beryllium window. For γ-ray densitometer, the intensity of the γ-ray beam was detected by another NaI crystal detector (Teledyne, S-44-I/2). The dimension of the crystal were as follows: 5.08 cm thick and 5.08 cm in diameter. The two detectors could be switched for use with different sources. The photomultiplier of the detector was connected sequentially to a preamplifier, an amplifier and a single-channel analyzer, a rate meter, and a 186 IBM compatible personal computer. The rate meter has a selector and a 0-100 mV scale range.

(3) Positioning Table. Both the source holder and detector were affixed to either side of the bed on a movable frame and could be moved anywhere up-or-down or to-and-fro by means of an electric motor.

c. High Resolution Micro-Imaging / Measuring System. The digital camera technique used to measure particle velocities as shown on Figure 4 comprised of the following units:

(1) Image Recording and Displaying Devices. A high resolution color video camera equipped with electronic shutter speed settings ranging from OFF to 1/10000 sec and super fine pitch color monitor were used to record and display solid velocities.

(2) Data Recording Device. A 486 / 33 MHz IBM compatible personal computer with a micro-imaging board inside and a micro-imaging software, Image-Pro Plus were used to record and store raw solid velocities data at any given location inside the fluidized bed.

d. Brookfield Viscometer. Brookfield digital viscometer (model LVDV-II+) with spring torque of 673.7 dyne-cm was used to measure the effective bed viscosities. This viscometer can produce twenty different rotational speeds ranging from 0 to 100 revolutions per minute (rpm) at four different modes, namely, LV, RV, HA and HBDVII+. The viscometer readings were recorded with a Hewlett-Packard (HP) LaserJet series II printer.

Experimental Procedure and Interpretation.

a. Fluidization Experiments. The liquid from the storage tank was fed to the bed from the bottom of the bed using the centrifugal pump. The gas was fed to the bed through a compressor. Both gas and liquid from the top of the bed were directed through three openings of 1.0 inch (2.54 cm) diameter back to the storage tank, where the gas was separated from the liquid.

In order to achieve a uniform fluidization, the liquid distributor section was designed in such a way that the pressure drop through the distributor section was 10 - 20 % of the total bed pressure drop. The gas was distributed in the fluidized bed through the six staggered porous tubes.

Air and water were used as the gas and liquid, respectively, in this experiment. Ballotini (leaded glass beads) with an average diameter of 0.8 cm and a density of 2.94 g/cm^3 were used as the solids. The experimental operating conditions are shown in the Table 4 (Bahary, 1994).

The experimental volume fractions measurements were made at 240 different locations at every 1.27 cm in both x and y -directions on the left portion of the bed. The experimental operating conditions are shown in the Table 4.

TABLE 4. Operating Conditions for Bubbly Coalesced Regime Experiments

Temperature (°C)	23.5
Particle Mean Diameter (cm)	0.8
Particle Density (g/cm^3)	2.94
Initial Bed Height (cm)	22 / 24
Minimum Fluidization Velocity (cm/s)	0.76

b. Volume Fractions Calibration. x-ray and γ-ray densitometers have been used to measure porosities of fluidized beds (Miller and Gidaspow, 1992: Seo and Gidaspow, 1987; Gidaspow, et al., 1983) and solids concentrations in nonaqueous suspensions (Jayaswal, et al., 1990). These techniques are based on the fact that the liquid, gas and solid phases under consideration have different absorptivities for x-ray and γ-ray. The same concept was adopted to measure concentration profiles inside our three phase fluidization systems.

The intensity of the transmitted x-rays or γ-rays can described as a function of the volume fractions of liquid, gas and the solid phases. The amount of radiation that is absorbed by a material can be given by the Beer-Bougert-Lambert Law:

$$I = I_o \exp(-krl) \tag{1}$$

where I is the intensity of transmitted radiation, I_o is the intensity of incident radiation, κ is the attenuation coefficient, ρ is the density of material, and l is the path length. The attenuation coefficient is calculated from a calibration curve. The logarithmic form of equation (1) is

$$\ln\left(\frac{I_o}{I}\right) = A \tag{2}$$

where $A=\kappa\rho l$.

For three phase (gas-liquid-solid) fluidized beds, the intensity of the transmitted x-ray or γ-ray could be described as a linear function of the volume fractions of the phases as:

$$\ln\left(\frac{I_x}{I_{o,x}}\right) = A_{g,x}e_g + A_{l,x}e_l + A_{s,x}e_s \tag{3}$$

$$\ln\left(\frac{I_g}{I_{o,g}}\right) = A_{g,g}e_g + A_{l,g}e_l + A_{s,g}e_s \tag{4}$$

where I_x and I_γ are the intensity readings of the x-ray and γ-ray densitometers; and ε_g, ε_l, ε_s are the volume fractions of gas, liquid and solid phases, respectively. The relation for volume fractions is:

$$e_g + e_l + e_s = 1 \tag{5}$$

The coefficients in equations (3) and (4) were calculated using the least square error technique from the calibration measurements of the intensity readings of x-ray and γ-ray densitometers at known concentrations of gas, liquid and solids in three phase mixtures. However, these coefficients were found to have values with 20% of error for x-ray and 2% of error for γ-ray.

To obtain the time averaged volume fractions at a designated position inside the three phase fluidized bed, an integration time of 40 seconds was used. The volume fraction measurements were obtained after a set of linear calibration was completed for x-ray and γ-ray densitometers as shown in Figure 6. From calibration curves, the time average values of volume fraction for liquid, gas and solid phases were calculated.

c. Velocity Measurements. In order to get a good visualization of microscopic movement of particles, a fiber-optic light was reflected on the field of view in the front and the back of the bed. The field of view in most experiments was a 2 cm × 2 cm area. As the particles were fluidized inside the bed, the camera with a zoom lens 18-108 mm and close up focus transferred its field of view to the monitor with streak lines. These streak lines represented the space traveled by the particles in a given time interval specified on the camera. The images were then captured and digitized by a micro-imaging board and analyzed using Image-Pro Plus software. Radial and axial velocity measurements were conducted at different locations inside the bed. The velocity vector was calculated as,

$$v_x = \frac{\Delta L}{\Delta t}\cos(a) \tag{6}$$

$$v_y = \frac{\Delta L}{\Delta t}\sin(a) \tag{7}$$

where, ΔL is the distance traveled, α is the angle from radial axis, Δt is the inverse of shutter speed, and v_x and v_y are the axial and radial velocity components, respectively.

d. Viscosity Measurements using Brookfield Viscometer. The viscometer was placed at the top of the fluidized bed and secured over the centerline of the bed. A cylindrical spindle (#1 LV) of 0.9421 cm diameter, 7.493 cm effective length and overall height of 11.50 cm was used. The cylindrical spindle was attached to the bottom of the viscometer without the guard and was lowered inside the fluidized bed by an extension wire until it was completely immersed in the mixture during measurements.

The measurements in this experiment were made under LV mode at different speeds between 2 and 20 rpm. At each rotational speed, between 10 and 30 readings were taken. The calibration of the viscometer-spindle apparatus was done using a Newtonian liquid, namely, water, using the procedure of Hetzler and Williams (1969).

The solids viscosity was calculated from measured ("apparent" or effective) homogeneous bed viscosity in fluidized bed defined as,

$$m_{bed} = e_g m_g + e_l m_l + e_s m_s \tag{8}$$

However, the effect of liquid and gas viscosities is negligible. In liquid-solid fluidized beds, gas volume fraction (ε_g) is zero.

e. Instantaneous Velocity Fluctuation and Granular Temperature. For Maxwellian distribution of velocity, mean axial and radial velocity components can be calculated as the

arithmetic averages. The variances of axial and radial particle velocities (following Maxwellian distribution) can be calculated from their mean values using

$$\left\langle C^2 \right\rangle = \frac{1}{N-1}\sum_{i=1}^{N}\left(v_i - v_m\right)^2 \qquad (9)$$

where, v_i and v_m are the i-th instantaneous and mean particle velocities, respectively.

The two-dimensional fluidized bed used in the experiment allows flows predominantly in two directions, namely axial (y-) and radial (x-). Hence. the instantaneous velocities measurements were made in axial-radial (y-x) plane only. However, the depth (z-) direction also has a non-zero component of particles fluctuations because of a finite bed depth. It is reasonable to assume that the $\left\langle C_z^2 \right\rangle = \left\langle C_x^2 \right\rangle$, because both z- and x- directions have zero mean gas and liquid velocities. Thus, the velocity fluctuations can be written as:

$$\begin{aligned}\left\langle C^2 \right\rangle &= \left\langle C_x{}^2 \right\rangle + \left\langle C_y{}^2 \right\rangle + \left\langle C_z{}^2 \right\rangle \\ &= 2\left\langle C_x{}^2 \right\rangle + \left\langle C_y{}^2 \right\rangle\end{aligned} \qquad (10)$$

where $\left\langle C_x^2 \right\rangle$ and $\left\langle C_y^2 \right\rangle$ are the measured mean square velocity fluctuations in -x and -y directions, respectively. The granular temperature Θ_s , is related to the square of mean fluctuating velocity of particles $\langle C^2 \rangle$ as.

$$\Theta_s = \frac{1}{3}\left\langle C^2 \right\rangle = \frac{2}{3}\left\langle C_x{}^2 \right\rangle + \frac{1}{3}\left\langle C_y{}^2 \right\rangle \qquad (11)$$

f. "Apparent" Bed Viscosity from Granular Temperature. In two and three phase fluidized beds, the rheological behavior of the bed predominantly reflects the granular viscosity of the solids. Lun, et al. (1984) derived an expression for the viscosity. Gidaspow's (1994) similar expression is used here.

$$m_s = \frac{5 r_s d_p \left(p\Theta_s\right)^{\frac{1}{2}}}{48(1+e)g_o}\left[1+\frac{4}{5}(1+e)g_o e_s\right]^2 + \frac{4}{5}e_s^2 r_s d_p (1+e) g_o \left(\frac{\Theta_s}{p}\right)^{\frac{1}{2}} \qquad (12)$$

The radial distribution function, g_o , is given in section (4A) in Table 1. The granular temperature Θ_s , is either obtained from the kinetic oscillation energy balance, or by velocity measurement experiments.

Experiments. Results For Bubbly Coalesced Regime.

Phase Hold-Up. Figures 7(a), 7(b) and 7(c) show the experimental results in our two dimensional bed for the time-averaged volume fractions of the liquid, gas and solid, respectively. Figures 7(b) and 7(c) show that there exists a region of a maximum value of volume fraction of gas, slightly shifted away from the center line, and has a value of approximately 0.6. This region can be interpreted as a region through which most of the coalesced bubbles pass. Figures 7(a) and 7(c) show that there are few solids close to the wall. This is due to the downward flow of the gas and the liquid near the wall, causing solids to entrain and move down in that region.

Flow Pattern. The flow patterns inside the gas-liquid-solid fluidized bed were observed visually and by video recording. Figure 8(a) shows a typical photograph of flow pattern in the bubbly coalesced regime. There is downward flow at the wall, upward flow close but not quite at the center and bubble coalescence. The small bubbles estimated to be about 1 mm coalesce together to form the large bubbles of a 4 cm effective diameter. The correlation by Fan (1989) gives diameters of 2 to 4 cm. The flow pattern and also positions of two coalesced large bubbles are not symmetric with respect to the vertical center line of the bed. Figure 8(b) shows that the computed particle volume fractions, at least roughly, agree with visual observations.

Instantaneous Velocity Distribution. The measured velocity data were analyzed using frequency distribution plots. The frequency distribution plots for particle axial and radial velocities are shown in Figures 9(a) and 9(b) for three phase fluidized bed. Figures 10(a) and 10(b) show axial and radial velocities fluctuation distribution in a two phase fluidized bed. These figures clearly demonstrate that the velocity fluctuations in two and three phase fluidized beds follow approximately Maxwellian distribution. Hence, the basic assumption used to derive the kinetic theory of granular flows applies to fluidized beds.

Granular Temperature. Figures 11 shows granular temperature, calculated using particle velocity measurements, as a function of horizontal distance from centerline of the bed at two different heights. Granular temperatures shows a maxima near the center at the bottom of the bed.

The temperatures were calculated to be in the range of 100 to 1000 (cm/sec^2). The fluctuating velocity is in the range of 17 to 39 cm/sec (0.17 to 0.39 m/s). Typical values of the fluctuating velocities for gas-solid fluidized beds were computed by Ding and Gidaspow (1990) to be in the range of 0.17 to 1.7 m/s. Also, based on solids discharge rates from mass flow hoppers, Gidaspow et al. (1986) obtained critical granular velocities in the range of 56.39 to 113.49 cm/sec (0.5639 to 1.1349 m/s) for 86 to 1550 μm size glass beads and 105.19 cm/sec (1.0519 m/s) for 603 μm sized sand particles.

The mean fluctuating velocities obtained from bed viscosity measurements using the Brookfield viscometer are very close to those calculated based on the direct velocity

measurement experiments. Figure 12 shows a comparison of the measured and the calculated bed viscosities in fluidized beds with a uniform distributor. The figures show that there is reasonable agreement between the solids viscosity measured using the Brookfield viscometer and those calculated from granular temperature using kinetic theory and direct measurement of solids velocity using the high resolution micro image / measuring system technique.

Computational Results For Bubbly Coalesced Regime.

The three phase hydrodynamics model shown in Table 1 was used in the computations. As an initial condition, the gas-liquid-solid bed was assumed to be at the minimum fluidization state. The computer simulations were carried out in two different modes: symmetric and asymmetric. In the symmetric mode, the fluidized bed behavior was assumed to be symmetric across the centerline parallel to the axial direction, and the gas at the inlet was distributed uniformly. In this mode only half of the bed was simulated. In the asymmetric mode, it was assumed that the gas at the inlet entering the two sides of the bed had different volume fractions. This assumption was supported by visual observation of the experiment shown in Figure 7(a).

In the symmetric mode, the computations were carried out using a mesh of 1040 finite difference computational cells with 0.635 cm $\times$ 2 cm cell sizes. In the asymmetric mode, the complete bed was simulated. A total of 1504 computational cells with 1.016 cm $\times$ 2 cm cell sizes were used. The inlet and initial conditions for both the cases are shown in Figure 13. In both the modes, simulations were carried out for 20 seconds with time increment of 5×10^{-5} seconds. A no-slip condition was used at the walls. The computations were conducted on a Hewlett-Packard 730 workstation.

Figures 14(a) and 14(b) show contours for instantaneous volume fractions and flow patterns simulated using symmetric and asymmetric modes, respectively. Computed flow pattern in asymmetric mode correctly shows upflow in the center region as seen in the experiments (see Figure 8(a)). However, the symmetric mode simulation incorrectly shows downflow in the center region. For asymmetric mode, the computed time-average volume fraction contours inside the bed for liquid, gas and solids are shown in Figures 15(a), 15(b) and 8(b), respectively.

For both symmetric and asymmetric modes, gas bubbles have a tendency to coalesce midway between the center of the bed and the walls. In the asymmetric mode computation the computed gas bubble moves in a snake-like fashion as observed in the experiment. Also in the asymmetric mode the two large bubbles coalesce at the center of the bed while in the symmetric mode the two large voids do not coalesce.

For asymmetric mode, the computed velocity vectors inside the bed for all three phases are shown in Figures 16. The computed velocity is very high in the region of the large bubbles. Figures 8(a) and 16 show that all three phases have downward movement near

the wall and upward movement at the center, the same flow pattern also reported by Chen, Reese, and Fan (1994).

This downflow produces a rotation or particle vorticity, given by inviscid equation below,

$$\frac{\partial z}{\partial t} + v_x \frac{\partial z}{\partial x} + v_y \frac{\partial z}{\partial y} = -\frac{g}{\left(1 - e_{mf}\right)} \frac{\partial e_g}{\partial y} \tag{14}$$

The equation shows that the vorticity produces a change in gas volume fraction ε_g. Then although we supply gas at a uniform rate, the gas is no longer uniformly distributed. Furthermore, Bouillard and Gidaspow (1991) have shown that when the gas at the bottom moves faster than in the rest of the bed, there is a catch-up effect and large gas voids may form, as depicted by Figure 8(a). It is the packing effect of the bubbles that produces this regime. When the injected bubbles are far apart, for low gas velocities, the bubbles move up at constant velocities. In this regime the drag is independent of the gas volume fraction. It is given by the corrected Stoke's law. At high gas velocities, the gas volume fraction is high and we use the Ergun equation to compute the drag in our model. Here the paths of the small injected bubbles intersect and bubbles coalesce. The situation is identical to that described in chapter 6 for gas-solid fluidized beds by Gidaspow (1994).

Comparison between Experimental and Computed Results in Bubbly Coalesced Regime

Phase Hold-up. Computed time average solid volume fraction contours inside the bed are shown in Figure 8(b). A photograph of the experiment in Figure 8(a) shows that the solid is concentrated in the lower 25 cm portion of the bed. There is a murky interface between the solid and the liquid-gas in the upper portion of the bed. This is in part reality and in part caused by inadequate time averaging. Time averaging of simulated values was done over about 20 seconds. Averaging over a longer time period could not be used due to a small loss of particles observed both in the experiment and more so in the simulation. The computation has higher particles loss because walls effects were neglected in two dimensional formulation. To correct this deficiency a three dimensional code is under development. Figure 15(b) shows that there is more gas in the lower portion of the bed (in the presence of solids) than in the upper portion where only gas and liquid are present.

Figures 17 and 18 show comparison between the experimentally measured and the simulated time-average volume fractions for gas and solids phases at different radial positions inside the bed. The figures show that there is a good agreement between the experimental and the computational results near the center and the wall regions.

Flow Pattern. Figure 19 shows a comparison between experimental and computed time-averaged axial solids velocity profiles at a height of 13.0 cm from the bottom of the bed for asymmetric mode simulation. The figure shows good comparison between measured

and asymmetric mode computed velocity profiles. However, at a height of 2.54 cm, the residual effects of liquid inlets can be seen.

Particulate Viscosity. The variance obtained from data such as depicted in Figure 15 gives the granular temperature (Gidaspow, 1994). The "apparent" bed viscosity was calculated from granular temperature using kinetic theory model shown in part (4A) of Table 1. A comparison of the estimated bed viscosity and those measured using a Brookfield viscometer (Bahary, 1994) is shown in Table 5.

"Apparent" Bed Viscosity and Granular Temperature. The "apparent" bed viscosities are shown in Table 5. In this table, the liquid volume fractions are estimated using correlation (Kim, et al., 1972) shown in equations (1) and (2) and solid volume fractions are calculated from visually measured expanded bed heights under fluidizing conditions. The reported volume fractions are averaged values. The agreement is excellent.

Bubble Coalescence Criterion. In a slurry bubble column three basic regimes can be observed (Shah, et al., 1982; Fan, 1986). For a large column diameter at low gas velocities, which means at low inlet gas volume fractions in the proposed model, the bubbles are so far apart that their velocities, called C below and in Gidaspow's book (1994) in chapter 6, can be given approximately by a Stokes' type equation (15), shown below. Table 1, Eq. (5) shows the more general case.

$$C = \frac{\Delta \rho g d_{bubble}^2}{18 \mu} \tag{15}$$

In this dispersed bubble regime the small bubbles move up the column without coalescing. As the velocity increases a packing effect makes itself felt. Then as a special case of the relations given in Table 1, C can be given by the first term of the Ergun equation for a packed column, shown in Equation (16).

$$C = \frac{\Delta \rho g d_{bubble}^2}{150 \mu_{mixture}} \varepsilon^2 = (\Delta \rho g)^{\frac{1}{3}} \frac{\left(6 \sigma D_{orifice}\right)^{\frac{2}{3}}}{150 \mu_{mixture}} \varepsilon^2 \tag{16}$$

In the second part of equation (16), the bubble diameter has been replaced by its expression obtained from its formation at an orifice at the distributor. This formula shows the effect of surface tension. A different way to compute the bubble diameter is to use a hydrodynamic stability analysis. In this study a constant bubble diameter was used. Hence surface tension enters the analysis only indirectly through bubble diameter.

TABLE 5. Three-Phase Fluidized Bed Solids Viscosity Comparison.

Sup. Gas Velocity (cm/s)	Sup. Liq. Velocity (cm/s)	Solid Vol. Fraction	Granular Temperature $(cm/s)^2$	Viscosity from Velocity Measurements	Viscosity from Brookfield Viscometer
3.364	2.037	0.346	845	4.26	4.36
3.364	4.027	0.272	435	2.95	3.13

The square dependence on porosity in Equation (16) gives the mechanism for bubble coalescence. At the inlet the porosity is larger than in the upper portion of the bed, hence the bubbles entering the bed move faster than those in the upper portion of the bed. There is a catch-up effect and hence bubbles coalesce. This is a possible explanation for the presence of small and large bubbles in the bubble coalesced regime. When the column diameter is small these large bubbles coalesce into a slug.

PART III: GRANULAR TEMPERATURE AND REYNOLDS STRESS FOR METHANOL CATALYST

Slurry Bubble Column Reactor

Slurry bubble column reactors have recently (Parkinson, 1997) become competitive with traditional tubular fixed-bed reactors for converting syn-gas into liquid fuels. In the U.S. a plant is being built by Air Products and Eastman Chemical (DOE, 1997) to produce methanol from syn-gas in a slurry bubble column reactor.

The advantage of a fluidized bed reactor over that of a fixed bed reactor is better heat and mass transfer due to constant agitation of the catalyst and the ability to introduce and remove the catalyst into the reactor. Such an operation requires an understanding of the flow of the catalyst.

Except for proprietary work of EXXON and other companies, the design of slurry bubble column reactors for indirect liquefaction is being done by the use of one-dimensional models. All hydrodynamic input is through empirical holdup correlations. The state of the art of the design of the slurry bubble column reactors is illustrated by the Viking Systems International (1993) report to PETC, now reorganized as the Federal Energy Technology Center (FETC).

The model presented in this paper permits the computation of three phase hydrodynamics. The only empirical input needed is an effective restitution coefficient of the particles (Gidaspow, et al., 1995; Wu, 1996; Pape, et al., 1996).

A 2-D THREE-PHASE BED WITH MULTIPLE JETS

Bed structure

To simulate a slice of the Air Products methanol reactor (Air Products, 1991) with jets a thin rectangular three phase fluidized bed was designed and constructed.

In order to have a symmetric system seven identical jets (7/8×7/8×7/8 inches) are installed at the bottom of the bed. See Figure 20. Each of these jets is connected to a valve to control the gas flow rates . The reason for using square jets is to eliminate the 3D effects caused by circular jets. At the bottom of each jet a cloth type material (with a pressure drop of 2 psi at the maximum gas flow rate) is used to prevent the 50 μm catalyst particles from going inside the gas pipe. The bed is constructed of 1/2 inch thickness Plexiglas. An opening at the top of the bed lets the air go out of the bed to the atmosphere.

The air flow rate can be varied up to 190 cf/hr at a pressure of 40 psi in order to achieve a superficial gas velocity of 12 cm/sec. In a typical Air Products experiment the weight ratio of solid to liquid was about 2:3. Figure 21 shows bed size and measurement location.

Granular Temperature and Reynolds Stress Measurements

The influence of the solid concentration on the granular temperature has been studied. For each run the solid-liquid ratio has been changed by loading more solids in the bed. Due to the small particle size a uniform dispersion is assumed to exist in the experiment. With this assumption the average bed expansion has been calculated and in this manner the solid volume fractions were obtained. In all of the experiments the superficial gas velocity was kept constant and all of the measurements were conducted at the same location of the bed so that the only variable is the solid concentration.

The granular temperatures and Reynolds Stresses have been calculated from instantaneous particle velocity measurements. These instantaneous particle velocities have been measured using a series of processes utilizing different tools. A CCD camera and built-in hardware were used to capture the trace of particles moving inside the bed. At this stage the illumination technique and also the camera speed play a significant role to get a sharp image. The captured particle traces were digitized with the aid of an advanced image processing software (IPPLUS®). Necessary operations such as background correction and filtrating were performed. The final image was used to measure the length and horizontal angle of the streak lines in order to obtain vertical and horizontal velocities. The histograms were drawn and the variance of the velocities have been calculated. See Fig 22 for a sample of a histogram. In part II of this study the granular temperature was calculated using the assumption that the velocity distributions were Maxwellian. Since such an assumption is too rough an approximation, in this section the granular temperatures were calculated using the method of Gidaspow and Huillin (1996) applied to FCC particles in a riser. The granular temperature was calculated using the equation for variance of velocity in the i direction :

$$s_i^2 = \frac{1}{N}\sum_{i=1}^{N}\left(v_i - v_m\right)^2 \qquad (17)$$

and the assumption of equality of the variances in the non-flow (z and x) directions:

$$q = \frac{1}{3}\left(+ s_{y}^{2} + s_{z}^{2}\right) = \frac{2}{3} s_{x}^{2} + \frac{1}{3} s_{y}^{2} \qquad (18)$$

The Reynolds stress has been calculated using the following formula:

$$\overline{u'v'} = \frac{1}{N}\sum \left(v - \bar{v}\right)\left(u - \bar{u}\right) \qquad (19)$$

Figure 23 shows the results of these experiments. As can be seen the granular temperature passes through a maximum value at low solids loading around ε_S=0.01. In the dilute limit as shown by Gidaspow(1994), the equation for the granular temperature, that is the balance of random kinetic energy, shows that the granular temperature θ is proportional to the volume fraction of solids, ε_s raised to the 2/3 power. This is analogous to the increase of thermal temperature of ideal gases with density upon compression, where the 2/3 corresponds to the ratio of the specific heats. Classical kinetic theory of granular flow gives an infinite granular temperature at zero solid volume fraction for shear flow. For high volume fractions of solids, Figure 23 shows the classical decrease of the granular temperature with volume fraction. This is known to be due to the decrease of the mean free path. For packed beds there will be no particle oscillations. Hence the behavior of the granular temperature in Figure 23 is general and is not restricted to the flow of 75 μm FCC catalyst in the IIT CFB or to Air Products catalyst in our unit. Operationally this means it may not be desirable to operate at very high catalyst concentrations due to reduced stirring at the high catalyst concentrations. This behavior is very similar to that observed by Gidaspow and Huillin (1996) in a circulating fluidized bed of 75 μm FCC particles.

We have observed much higher granular temperatures with gas flow than was the case for liquid flow with Air Products catalyst. This means there is much better stirring caused by the high gas flow; clearly a desirable feature in a reactor. There the highest granular temperature was only one $(cm/sec)^2$, one hundred times lower .Hence the high gas flow produces the desirable stirring .

A Link Between the Granular Temperature and the Particle Reynolds Stress

For steady, developed shear flow (Gidaspow, 1994; Cao and Ahmadi, 1995) with a negligible interaction energy supply, i.e. equal particle and fluid oscillations, the production of oscillations by particle collisions equals the dissipation due to inelastic collisions. With these approximations the fluctuating kinetic energy equation (Eq. (4A) in Table 1) reduces itself to the balance:

$$-P_{xy}\cdot\frac{\partial u_s}{\partial y}=12\,e_s^{\,2}\,\rho_s g_o\left(1-e^2\right)\left(\frac{q}{d_p}\right)\left(\frac{q}{p}\right)^{\frac{1}{2}} \tag{20}$$

production of particle oscillations by shear P_{xy} = dissipation due to particle collisions with an effective restitution coefficient , e

where u_s is the particle velocity in the x direction, the basic flow direction, and y is the perpendicular direction.

Gidaspow (1994) used such an equation to estimate the restitution coefficient in a CFB where the shear rate, $\left(\frac{\partial u_s}{\partial y}\right)$ was known. Here the shear, P_{xy} was directly measured.

From its definition,

$$P_{xy}=\rho_s e_s\left\langle C_x C_y\right\rangle \tag{21}$$

where C_i is the instantaneous minus the average particle velocity in the i direction and $\langle\ \rangle$ means averaging over the velocity space. In terms of the conventional turbulence theories, Equation (19) gives the particle Reynolds stress

$$P_{xy}=\rho_s e_s\overline{u_s' v_s'} \tag{22}$$

where the bar over the velocities has the same meaning as the bracket averaging.

Equations (20) and (22) provide a link between the Reynolds stress and the granular temperature . This relation involves the as yet unknown shear rate. The shear rate can be eliminated using a second expression for shear stress.

$$-P_{xy}=\mu_s\frac{\partial u_s}{\partial y} \tag{23}$$

where μ_s is the viscosity of the solid.

For the dense flow, this viscosity is given by

$$\mu_s = \frac{4}{5} \varepsilon_s^2 \rho_s d_p g_o (1+e) \left(\frac{\theta}{\pi} \right)^{\frac{1}{2}} \tag{24}$$

As a limiting value the balance between production of oscillations and their dissipation, as given by Equation (20), was previously (Gidaspow, 1994) shown to be

$$\left(\frac{1}{15} \right)^{\frac{1}{2}} \frac{\partial u_s}{\partial y} d_p = (1-e)^{\frac{1}{2}} \theta^{\frac{1}{2}} \tag{25}$$

This equation permits the elimination of the shear rate in Equation (20). Division of Equation (20) by (25) and use of (22) gives the link between the Reynolds stress and the granular temperature in terms of inelastic dissipation.

$$\frac{\overline{u_s' v_s'}}{3\theta} \equiv \frac{u_s' v_s'}{u_s'^2 + v_s'^2 + w_s'^2} = \left(\frac{16}{15\pi} \right)^{\frac{1}{2}} \varepsilon_s g_o \frac{(1+e^2)}{(1-e)^{\frac{1}{2}}} \tag{26}$$

For nearly elastic particles , Eq. (26) gives

$$\frac{\overline{u_s' v_s'}}{3\theta} = 1.1954 \varepsilon_s g_o (1-e)^{\frac{1}{2}} \tag{27}$$

which is valid for the dense regime.
The dilute limit yields an expression that is independent of particle concentration, as

$$\frac{\text{Reynolds Stress}}{\text{Granular Temperature}} = \frac{\overline{u_s' v_s'}}{\theta} = \left(\frac{5}{8} \right)^{\frac{1}{2}} (1+e^2)^{\frac{1}{2}} \quad \text{dilute limit} \tag{28}$$

For isotropic turbulence Eq. (28) gives no real solution for the restitution coefficient.

Particle Attrition

Air Products methanol catalyst particles that have been used as the solid phase in our three phase fluidization have been examined to find the breakage. The fresh catalyst was analyzed by a sieve method. The distribution of particle size is given in Figure 24.
After one year usage of the catalyst in the bed in which the air was always flowing at a velocity of at least 0.2 cm/sec through seven jets, the particles have been analyzed by a Reflected Light Microscope. The pictures from microscope were transferred to our image processing software (IPPLUS®) where the particles size were measured. Two pictures

that have been captured are shown in Figure 25. The distribution of particle size is given in Figure 26. As can be seen the used catalyst is mostly fines and much smaller than the original fresh catalyst. The distribution has a double peak. It shows that there was a lot of fine particles production due to jets and particle-particle and particle-wall collisions.

TABLE 6 - Restitution Coefficient for 50 μm Catalyst in G-L-S Bed

e_s	$\frac{\overline{u'v'}}{q}$	$1-e$
0.13	0.2873	0.06837
0.07	-0.4127	0.14707
0.02	-0.4069	0.14263
0.0075	-0.0716	0.00411
0.0004	0.2461	0.04969

Cluster Formation

As shown in Figure 27, some clusters are formed. These clusters are a group of particles and may be considered as one moving object. Indeed the viscosity of the Air Products catalyst slurry measured with a Brookfield viscometer was ten times that estimated using kinetic theory, based on 50 μm catalyst particle size.

Optimum Catalyst Design

The measurements reported here permit us to speculate concerning optimum Fischer-Tropsch catalyst design. The diffisusional resistance inside the catalyst leads one to make the catalyst as small as possible, since the resistance is proportional to particle diameter. However, this study shows that small particle diameters lead to small particle oscillations and to agglomeration. This gives a large particle resistance to mass transfer. Hence there exists an optimum particle size. Furthermore as seen from Figure 23 and theoretical speculations as the particle concentration increases, the particle oscillations decrease, stopping completely in the packed bed state. Hence there is also some optimum catalyst concentration.

The particle oscillations in Fig. 23 are much larger than for the liquid-catalyst system (Gidaspow and Huillin, 1997). They are caused by the flow of the gas. Although much information in this area proprietary, obtained by expensive trial and error of slurry reactor design, it appears that gas flow rates used in this technology are substantially above those needed for reaction and are used to promote stirring. This is another area needing modeling. The proposed CFD model was developed to answer such questions. Wu (1996) and Gidaspow, et al.,(ASME , 1996) have developed a reaction model.

CONCLUSIONS

1. Discussion at various National Science Foundation-Department of Energy (NSF-DOE) workshops in the late eighties and early nineties produced a general agreement between the participants that for validation of time averaged multiphase flow equations it is necessary to compare the computed phase velocities and concentrations, sometimes called hold-ups, to well defined experiments. In this study we measured the particle velocities with a new technique and hold-ups with a combination of an x-ray and a γ-ray densitometers. There is a fair agreement between the model and the experiments. The differences are principally due to non-uniformities in our distributor.

For 800 μm glass beads, computed and measured time average velocities agree in the sense that :

a. There exists down flow at the walls.
b. The velocities peak in the region of most frequent bubble motion.
c. In the center of the bed there is a small negative down flow.

2. Computations have shown the need for knowing phase viscosities. Kinetic theory shows that the viscosities can be obtained from granular temperatures. In this study we have developed a new technique for measuring the granular temperature of particles using a digital video camera. We have obtained an excellent agreement between the viscosities of a three phase and a two phase slurry obtained by measuring the granular temperature and by measuring the viscosity with a Brookfield viscometer.

3. The large value of the slurry viscosity, several poises for large particles, compared with the one centipoise viscosity for water clearly shows the dominant effect of particle collisions. In modeling this system we can ignore the liquid and the gas viscosities.

4. A link between the particle Reynolds stress and the granular temperature was developed for shear flow. It was demonstrated experimentally for Air Products Methanol catalysts.

5. From the measurement of the Reynolds stress and the granular temperature of a methanol catalyst in a three phase bubble coalesced fluidized bed with multiple jets the effective restitution coefficient was obtained to be about 0.9. This parameter is the only unknown in the kinetic theory model. Its knowledge permits the simulation of methanol reactor hydrodynamics.

6. The present data are consistent with the Radiation Particle Tracking turbulence intensity and particle velocity measurements of Larachi, et al. (1996). In their fluidized bed they used 3 mm glass beads, had a higher expanded bed height and used higher flow rates than in the present study. Due to these factors they had upflow in the center and downflow at the walls. In this study we also have downflow. We can achieve upflow in the center by increasing the flow rates and particle inventory as demonstrated for a bubbling gas-solid fluidized bed (Gidaspow, 1994). The granular temperature measured and computed in this study is essentially the sum of the radial and axial turbulence intensities reported by Larachi, et al. The model in its present state does not compute the individual turbulence intensities.

Reynolds stresses were measured in L.S. Fan's laboratory at Ohio State University (Reese, 1996) using a PIV technique, similar to that used here. The ratio of the absolute value of

the shear Reynolds stresses to the granular temperature, computed from reported axial and radial normal stresses, have the same trend, as predicted by equation (28).

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NOMENCLATURE

Abbreviation	**Term**
A	absorbance (equals $\kappa\rho l$)
C, c	fluctuating velocity of a particle
C_D	drag coefficient
d_k	characteristic diameter of particulate phase k
e	coefficient of restitution
G	solid compressive stress modulus
g	gravity
g_o	radial distribution function
I	intensity of transmitted radiation
[**I**]	unit tensor
L	length traveled
l	path length
N	number of particle velocity measurements
p	pressure
q	flux vector of fluctuating energy
Re_k	Reynolds number of phase k
t	time
u	velocity in the basic direction of flow
v	velocity in the perpendicular direction
v	velocity vector
X_k	reduced volume fraction $\left\| e_k / \left(e_k + e_m\right) \right\|$

Greek letters

α	angle of particle movement coefficient in particle-particle drag coefficient equation
β_{km}	interphase momentum transfer between phase k and phase m
γ	collision energy dissipation
ε_k	volume fraction of phase “k”
ε	void fraction
ζ	vorticity
$3/2\Theta_s$	fluctuating energy
κ	conductivity of fluctuating energy

	mass attenuation coefficient
μ	shear viscosity
μ_s	(shear viscosity $\times$ volume fraction) of solids
ξ	bulk viscosity
ξ_s	(bulk viscosity $\times$ volume fraction) of solids
ρ	density
τ	stress
Φ_s	viscous energy dissipation
ψ_s	particle sphericity

Subscripts

0	incident
bed	effective bed
dil	dilute phase
g	gas phase
k	phase k
i	for i-th instantaneous measurement
l	liquid phase
m	phase m
	mean
max	maximum
mf	minimum fluidization
s	solid phase
sup	superficial
t	terminal
x	x-ray
x	x- axial component
y	y- radial component
γ	γ-ray

Superscripts

T	transpose

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