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ON

HYDRODYNAMIC MODELS FOR SLURRY BUBBLE COLUMN REACTORS

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ABSTRACT

HYDRODYNAMIC MODELS FOR SLURRY BUBBLE COLUMN REACTORS

The objective of this investigation is to convert our "learning gas-solid-liquid" fluidization model into a predictive design model.The IIT hydrodynamic model computes the phase velocities and the volume fractions of gas,liquid and particulate phases.Model verification involves a comparison of these computed velocities and volume fractions to experimental values.

This report includes a paper submitted for review for presentaation at the Second International Symposium on Numerical Methods for Multiphase Flows,ASME Fluids Engineering Division,San Diego,CA,July 7-11,1996.We found a severe stability restriction that all codes with chemical reaction must satisfy to have meaningful results.This is explained in the paper.

Next quarter report will present our experimental measurements of granular temperature of Air Products catalyst.

MULTIPHASE FLOW IN SLURRY BUBBLE COLUMN REACTORS AND SOLID PROPELLLANT ROCKETS

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ABSTRACT

A two-dimensional,transient computer code for solving a generalization of Navier-Stokes equations for reacting multiphase flow was developed and tested for two applications:production of methanol in an Air Products slurry bubble column reactor and generation of particles in a rocket motor.

For the slow catalytic methanol production the conventional ICE technique produced numerical solutions in agreement with Air Products pilot plant results and IIT's hydrodynamics experiments.The code predicted the measured methanol producton,the observed vortices and the catalyst viscosity obtained from measurements of granular temperature using a digital camera.

However,for the rapid propellant combustion the conventional ICE technique proved inadequate.In such problems the absolute error grows without bounds for explicit and for implicit numerical schemes,as,for example, determined by von Neumann stability analysis.An analysis of the relative error showed how to finite difference the rate of reaction to obtain numerically stable solutions.

INTRODUCTION

For the past 20 years at IIT we have been developing the theory and computer codes for multiphase flow and fluidization.The recent version of the theory is based on the concept of granular temperature - the random oscillating kinetic energy-pioneered by professor Stuart Savage of McGill and collaborators[1] and on the generalization of the Navier-Stokes equations for multiphase flow.In this theory the particulate phase has its own equation of state,viscosity,etc.Gidaspow and Huilin have recently[2] verified that for a dilute particle gas mixture the particle pressure equals the bulk density times the granular temperatue.In other words the particulate phase obeys an analogue of an ideal gas law.The particulate viscosity was obtained from measurements of granular temperature.Gidaspow[3] reviewed this theory.

The computer codes we developed are based on the Los Alamos K-FIX code[4] which uses the ICE method of solution.Syamlal and others of Morgantown incorporated some of the kinetic theory into the commercial FLUENT code.Most of our research was without reaction.When we tried simulatons with rapid reaction we ran into numerical problems.This led us to perform the stability analysis reported in this paper.We believe this analysis should be useful for codes such as KIVA that are widely used for engine combuston simulations[5].

STABILITY ANALYSIS

General

A stability analysis has been conducted on the finite difference form of the continuity, momentum and energy equations. To illustrate the method, the continuity equation will be discussed. When considering only convective and reaction terms, the continuity equation is

$$\frac{\partial(\varepsilon_k \rho_k)}{\partial t} + \nabla(\varepsilon_k \rho_k v_k) = \dot{m}_k \qquad (1)$$

The finite difference form of the continuity equation in the IIT code is

$$(\varepsilon_k\rho_k)_{i,j}^{n+1} = (\varepsilon_k\rho_k)_{i,j}^{n} - \frac{\delta t}{\delta r_i} < (\varepsilon_k\rho_k)u_k >_{i,j}^{n+1} - \frac{\delta t}{\delta z_j} < (\varepsilon_k\rho_k)v_k >_{i,j}^{n+1} + \delta t \cdot \dot{m}_{k_{i,j}}^{n} \quad (2)$$

The mass generation term can be represented in the following form:

$$\dot{m}_{kij}{}^{n} = (\varepsilon_k\rho_k)_{ij}^{n}\ z_k\ \exp[-\frac{E_k}{RT_{kij}{}^{n}}] \quad (3)$$

To simplify the analysis, consider Cartesian coordinates, only the z-direction, and constant incremental distance. It is also assumed that all velocities are in the positive z-direction corresponding to a specific donor cell differencing configuration. Temperature is also taken as constant. In this case, the donor cell flux is given by:

$$\left\langle(\varepsilon_\kappa\rho_\kappa)v_k\right\rangle_j^{n+1} = v_{kj+\frac{1}{2}}^{n+1}(\varepsilon_\kappa\rho_\kappa)_j^{n+1} - v_{kj-\frac{1}{2}}^{n+1}(\varepsilon_\kappa\rho_\kappa)_{j-1}^{n+1} \quad (4)$$

Velocity will be taken as a constant, U, and the following parameters are defined:

$$F'_i = F_i^{n+1} = (\varepsilon_\kappa\rho_\kappa)_i^{n+1};\ F_i = F_i^n;$$

$$U = v_i = v_{k_i}^{n+1};\ \dot{m}_{k_i}^n = (\varepsilon_\kappa\rho_\kappa)_i^n M;\ R = \frac{\delta t}{\delta r}$$

With these substitutions, the continuity equation becomes $F_i = F_i - (RUF_i - RUF_{i-1}) + M\delta t F_i$ (5)

Although, this is the form of the conservation of mass equation in the code, the formulation could also be fully explicit or fully implicit as given below.

Fully Explicit Form:

$$F'_j = F_j - RU(F_j - F_{j-1}) + M\delta t F_j \quad (6)$$

Fully Implicit Form:

$$F'_j = F_j - RU(F'_j - F'_{j-1}) + M\delta t F'_j \quad (7)$$

Conservation of Momentum and Energy

In the IIT code, the conservation of momentum and energy equations are in mixed implicit - explicit form. With respect to terms considered here (convection and reaction) the momentum equation is fully explicit. For stability analysis we neglect pressure gradients, gravity, shear and drag terms in the momentum equation. In the energy equation, we neglect pressure gradients, convective heat transfer, conduction, viscous dissipation and gas particle drag dissipation. For momentum, $F_j = (\varepsilon_k \rho_k v_k)_{j+\frac{1}{2}}^n$ and M has identical meaning to that used in the continuity assessment. For the energy equation, $F_j = (\varepsilon_k \rho_k H_k)_j^n$ and M must be corrected by multiplying by the parameter $\eta = \Delta H/H_k$

Von Neumann Absolute Stability Analyses

It is recognized that all of the relations described above can be fully explicit, fully implicit or mixed form. Each of these forms is analyzed by applying a Von Neumann stability analysis. Separation of variables is accomplished by assuming F to be of the form:

$$F_j^n = \lambda^n\ e^{jl\theta} \quad (8)$$

Using this assumed form in the various model equations and solving for λ (the amplification factor), stability criteria are derived by observing that the amplification factor must be less than or equal to 1 for stability.

A classical Von Neuman absolute stability analysis [6] was applied to the three differencing equations. By this technique it is found that the fully explicit form requires that M be negative in order to have absolute stability in some regime of RU. With the implicit form, absolute stability imposes strange restrictions on Mδt, depending on the value of RU. For example, when RU is 0, Mδt must exceed 2 for absolute stability. The mixed form is found to have the restriction:

$$-2 \le M\delta t \le 0 \quad (9)$$

The implications of these results would be devastating, if true. In multiphase reactive flow modeling, whenever there is consumption of a phase (negative M) there is simultaneous generation in another phase (positive M). Negative M is not a problem, but absolute stability restrictions are a severe problem when M is positive. This is a direct consequence of the form of the equations. Analytic solution of the simplified equations for the duration of one time step yields:

$$F_j^{n+1} = F_j^n e^{M\delta t} \quad (10)$$

If time (or δt) were allowed to increase without bound, F_j^{n+1} would also be unbounded, and this is reflected in the amplification factor.

Relative Stability

Morton and Richtmeyer [6] point out that the absolute stability criteria is too restrictive for situations of the type described here. Instead, stability should be assessed relative to the desired analytic solution for the configuration of interest. As indicated above, the maximum change in F due to the reaction term will be

$$F_j^{n+1} = F_j^n\ e^{M\delta t} \quad (11)$$

This is the maximum change due to reaction, and it is the analytic solution used to assess relative stability. For assessment of relative stability, we have $|\lambda| \le e^{M\delta t}$ instead of $|\lambda| \le 1$, as was required for absolute stability.

Fully Explicit Numerical Scheme. For the fully explicit scheme, including reaction and donor cell

differencing with a positive velocity, the finite difference equation is

$$F'_j = F_j - RU(F_j - F_{j-1}) + M\delta t F_j \qquad (12)$$

A Von Neumann stability analysis leads to the following amplification factor for absolute stability.

$$|\lambda| = [1 + 2(R^2U^2 - RU - RUM\delta t) (1 - \cos(\theta)) + M^2\delta t^2 + 2M\delta t]^{1/2} \le 1 \qquad (13)$$

When relative stability is evaluated, we have:

$$\frac{|\lambda|}{e^{M\delta t}} \le 1 \qquad (14)$$

By plotting amplification factor as shown in equation 13 or 14 versus $M\delta t$ for different values of RU, the region of stability can be identified. Figure 1 is a plot of absolute and relative stability for the fully explicit scheme for the specific case of RU = 0. In all cases with the explicit scheme, absolute stability requires that Mδt be negative. When RU = 0, with relative stability all positive values of Mδt have amplification factors less than or equal to 1, and are therefore stable. If either absolute or relative amplification factor is less than or equal to 1 in a range, the range will be stable. Figure 2 maps the bound of stability in the RU versus M δ t plane. All values to the right of the curve are stable. The rectangle at the bottom has been selected as a practical bound. We choose the following bounds for computations:

$$RU \le 0.5 \qquad (15)$$

$$-1 \le M\delta t \le +1 \qquad (16)$$

Fully Implicit Numerical Scheme. For the fully implicit scheme, including reaction and donor cell differencing with a positive velocity, the finite difference equation is

$$F'_j = F_j - RU(F'_j - F'_{j-1}) + M\delta t F'_j \qquad (17)$$

A Von Neumann stability analysis leads to the following amplification factor for relative stability.

$$|\lambda| = \frac{1}{\sqrt{a^2 + b^2}} \le e^{M\delta t} \qquad (18)$$

where $a = 1 + RU(1 - \cos\theta) - M\delta t$

and $b = RU\sin\theta$.

Figure 3 is a map of the primary stable zone for the fully implicit scheme. Absolute stability is achieved for all negative values of Mδt, but there are severe problems when Mδt is near +1. Relative stability has this problem also. This is due to a singularity that occurs because of the -Mδt term in the denominator for the amplification factor. Singularities show up at other Mδt values for other values of RU. In all cases the implicit scheme has problems for positive reaction terms. Based on absolute stability, all negative reaction terms are acceptable.

Mixed Form. A third numerical scheme is considered. If the stability problems are due to a singularity caused by the term (-Mδt) in the amplification factor denominator, the reaction term should be kept in the numerator. This is accomplished by always accounting for reaction explicitly, regardless of the form of the overall scheme. Therefore, consider all terms implicit with the reaction accounted for explicitly:

$$F'_j = F_j - RU(F'_j - F'_{j-1}) + M\delta t F_j \qquad (19)$$

A Von Neumann stability analysis leads to the following amplification factor for relative stability.

$$|\lambda| \le \frac{|c|}{\sqrt{a^2 + b^2}} \le e^{M\delta t} \qquad (20)$$

where $a = 1 + RU(1 - \cos\theta)$,

$b = RU\sin\theta$

$c = 1 + M\delta t$

It can be shown that the maximum of the magnitude of the amplification factor is independent of RU. For negative reaction terms, absolute stability controls., and Mδt must be greater than -2 for stability. For positive reaction terms relative stability controls, and there is no restriction on positive reaction terms. This is shown in figure 4. The mixed form is the least restrictive of the numerical schemes with respect to Mδt. The primary conclusion, however, is that the reaction term should be handled explicitly, regardless of the form of the overall numerical scheme being employed.

INTERPRETATION OF RESULTS

The stability guidelines presented above are applied to two problems, and the implications discussed. The problems assessed here are: (1) a fluidized bed reactor, and (2) a solid propellant combustion chamber.

Fluidized Bed Reactor. Simulations of Synthesis of Methanol in Air Products and Chemicals' Slurry Reactors

The governing equations for the hydrodynamic model and the chemical reactions were reported by Gidaspow,et al (1995) [7]. Table 1 shows the reactor's operating conditions. Table 2 shows the material balances. Table 3 shows the comparison of the simulation and Air Products' RUN E8.1[8]. Figure 5 shows the comparison of methanol concentration profiles from IIT's hydrodynamic model, one-dimensional model and Viking's model [9]. The hydrodynamic model predicted much better results than the other two models did. Figure 6 shows a transient gas flow pattern in the reactor. Figure 7 shows the transient solid and gas volume fraction, thermal temperature, granular temperature and viscosity profiles. The blocks in Figure 6 and 7 represent heat exchangers.

The reaction rates and flow velocities are both extremely low in this system. For this case, the stability parameters can be summarized as

$\delta t = 10^{-4}$ s $RU = 8.02 \times 10^{-4} < 0.5$

$\delta x = 1.9$ cm $M = 1.54 \times 10^{-2}$ s^{-1}

U = 15.24 cm/s $|M\delta t| = 1.54 \times 10^{-6} < 1$

Clearly, both stability criteria RU and $|M\delta t|$ are easily satisfied in this problem.

Solid Rocket Combustion Chamber. The simulation of a solid rocket combustion chamber is an ongoing investigation, and the stability analysis has been conducted to provide the insight necessary for proper setup of the simulations being done. A preliminary result is shown in figure 8. A number of computer experiments have been done using this configuration. One series of runs helps to clarify the meaning of the stability criteria. These simulations all used one microsecond time steps and 1 cm cell dimensions. Velocities were low in this configuration, so the RU criteria was easily satisfied. Therefore, the results for three specified constant reaction rates could be compared.

Case	Excessive Rate	Critical Rate	Low Rate
M (sec^{-1})	10^7	10^6	10^5
M δt	10	1	0.1
Result	Immediate Failure	Ran 14 time-steps (borderline)	Did not Fail

Clearly, when the problem is carefully setup to be in a specific parameter range, the expected stability results can be produced. More generally, it is observed that problems that involve high reaction rates also tend to produce high localized velocities. Then, the RU criteria controls stability. Many times, a higher resolution numerical grid is also required for such problems, and stability requires correspondingly smaller time steps. For example, when velocities are supersonic, perhaps 10^5 cm/s, and a cell dimension is 0.01 cm, the time-step must be less than $5x10^{-8}$ s. From a practical standpoint, it is found that at least an order of magnitude smaller time-step values are really necessary to compensate for local velocity fluctuations, perhaps within the convergence procedure.

ACKNOWLEDGEMENT

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NOMENCLATURE

E—activity energy of reaction
$F = \varepsilon\rho$
H—enthalpy

$\dot{m}$—rate of production
$M = z_k \exp\left[-E_k / RT_{k_{ij}}^n\right]$

$R = \delta t / \delta r$
t— time
T—temperature
u—velocity in r direction
U=v
v—velocity in z direction

Greek letters:
λ—eigenvalue
ε—volume fraction
ρ—density

Subscripts:
i,j—cell index
k—phase k

Supscripts:
n—the nth time step

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8. Air Products and Chemicals, Inc., "Liquid Phase Methanol LaPorte Process Development Unit: Modification, Operation, and Support Studies", Draft Report to DOE for Contract No. DE-AC22-87PC90005 (1991).
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DISCLAIMER

Table 1 Reactor Operating Conditions:

2-D reactor	22.5"x320"
pressure	735.2 psig
temperature	250.3 °c
gas velocity	15.24 cm/s
catalyst density	3.011 g/cm³
catalyst diameter	50 μm
liquid density	0.70025 g/cm^3
gas type	CO-rich

Table 2 Material Balance:

	Inlet		outlet*	
	% mol	kgmol/hr	% mol	kgmol/hr
CO	51	87.9	51.18	74.8
CO_2	13	22.4	15.39	22.4
H_2	35	60.3	23.03	33.6
CH_3OH	0	0.0	9.20	13.4
N_2	1	1.7	1.18	1.7
$\overline{M}$ (kg/kgmol)	20.98		24.837	
F (kgmol/hr)		172.3		145.9
F (kg/hr)		3614		3625**

* Taken at the vapor-slurry interface.

** Time averaging flowrate.

Table 3 Comparison of Simulation and Air Products' RUN E-8.1:

	CO conv. (%)	gas holdup (%)	slurry height (inches)	total catalyst (kg)	CH_3OH (gmol/hr/kg)	net CH_3OH (TPD)
Simulation	14.24	26.9	215	740	16.93	9.62
RUN E-8.1	13.50	29.5	200	567	20.50	10.03

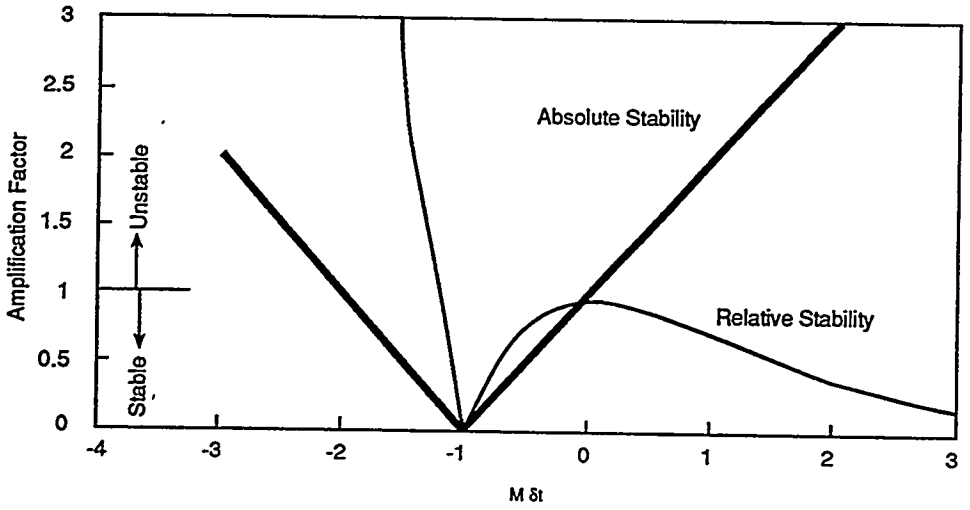

Figure 1. Stability For Explicit Form with Reaction and for RU = 0

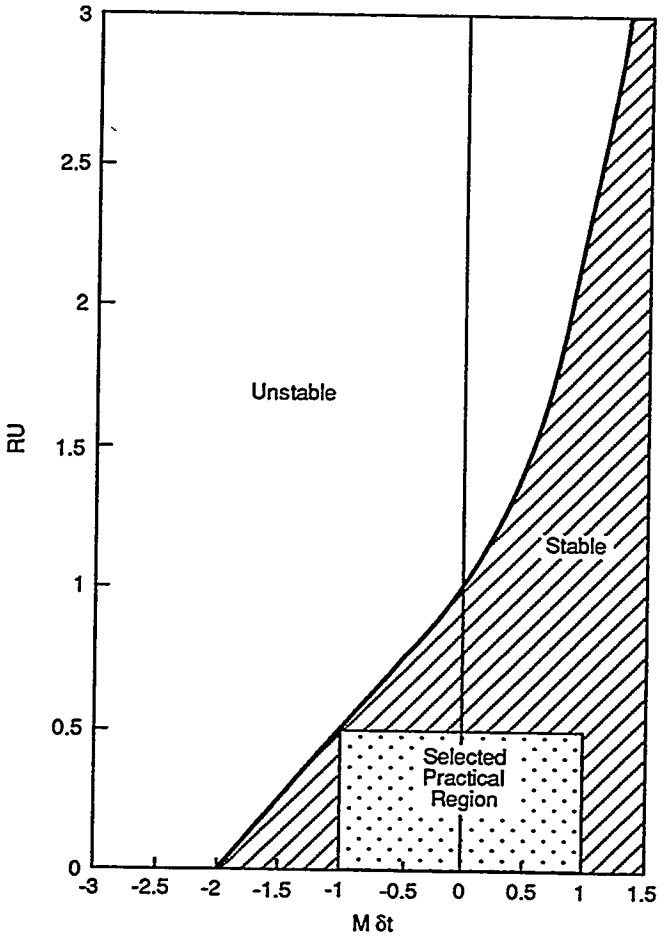

Figure 2. Stabillity Map for Fully Explicit Form

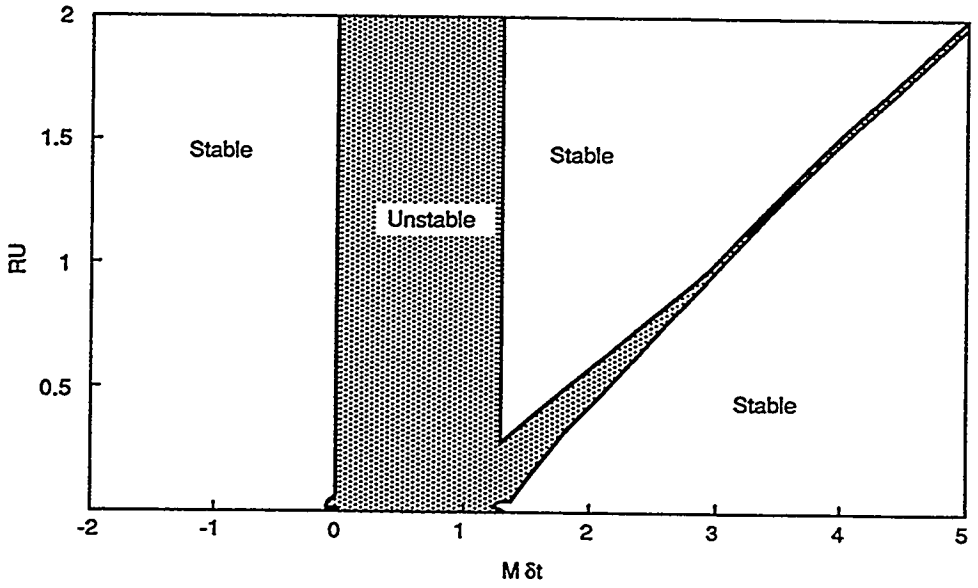

Figure 3. Stable Regions for Implicit Scheme with Reaction

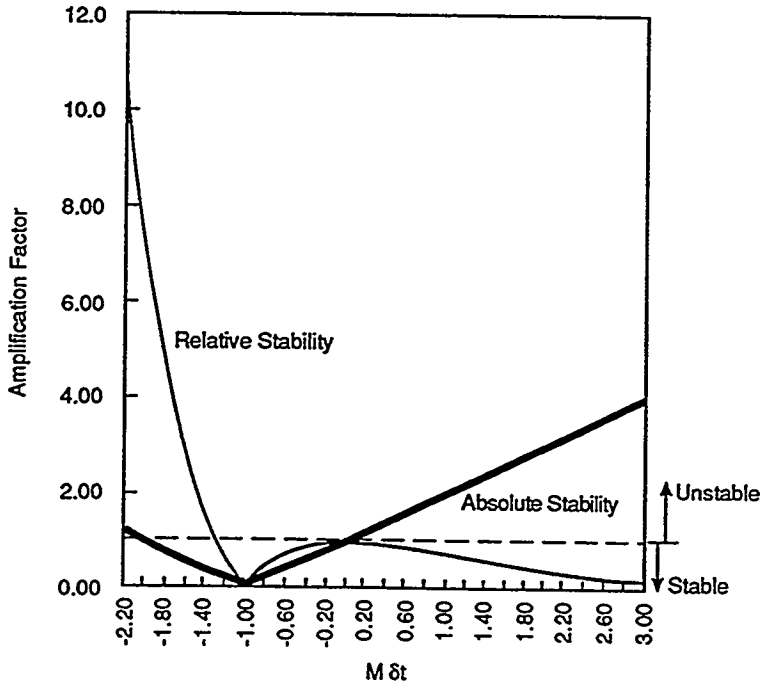

Figure 4. Absolute and Relative Stability for Mixed Form

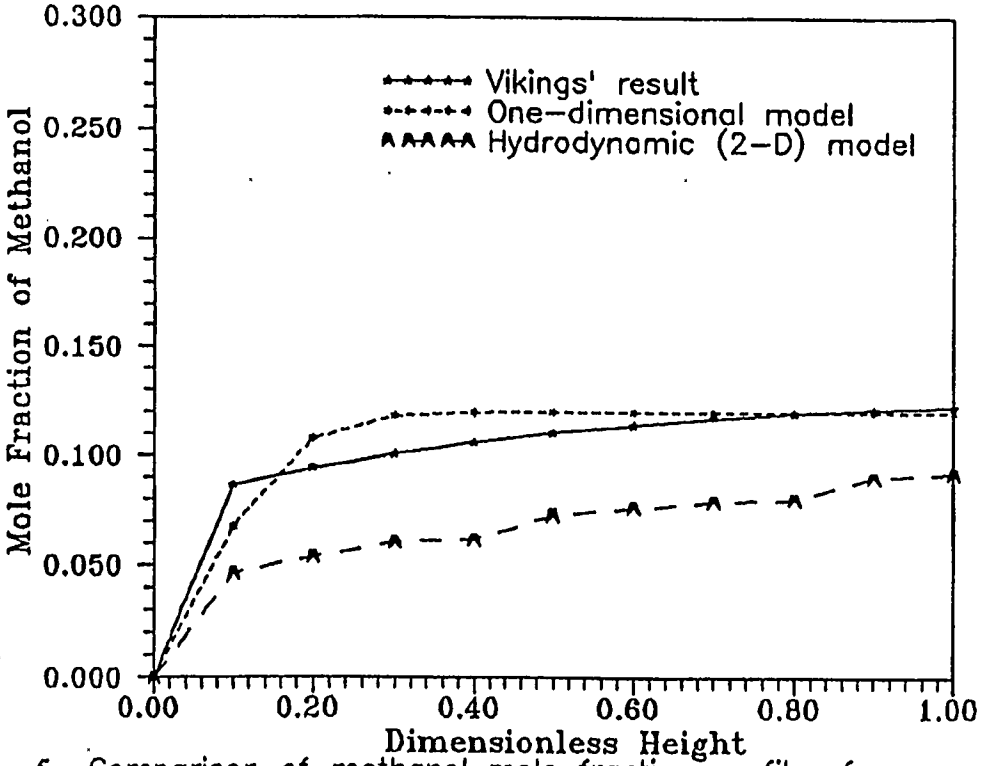

Fig. 5 Comparison of methanol mole fraction profiles from IIT's hydrodynamic model, one-dimensional model and Vikings' model.

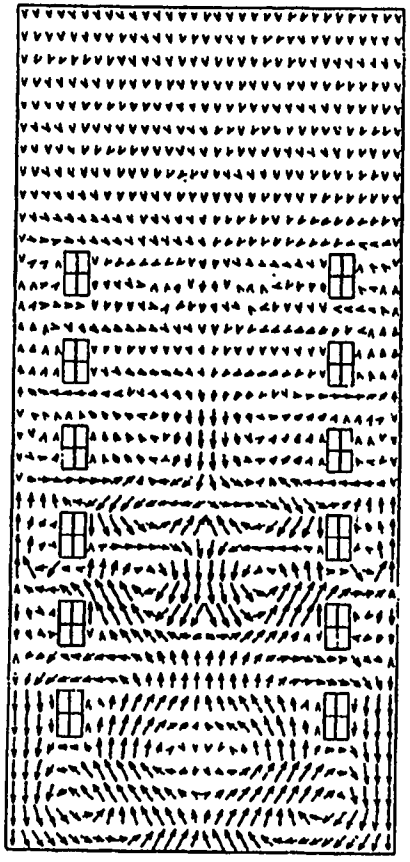

Figure 6 Computed Gas Transient Flow Pattern.

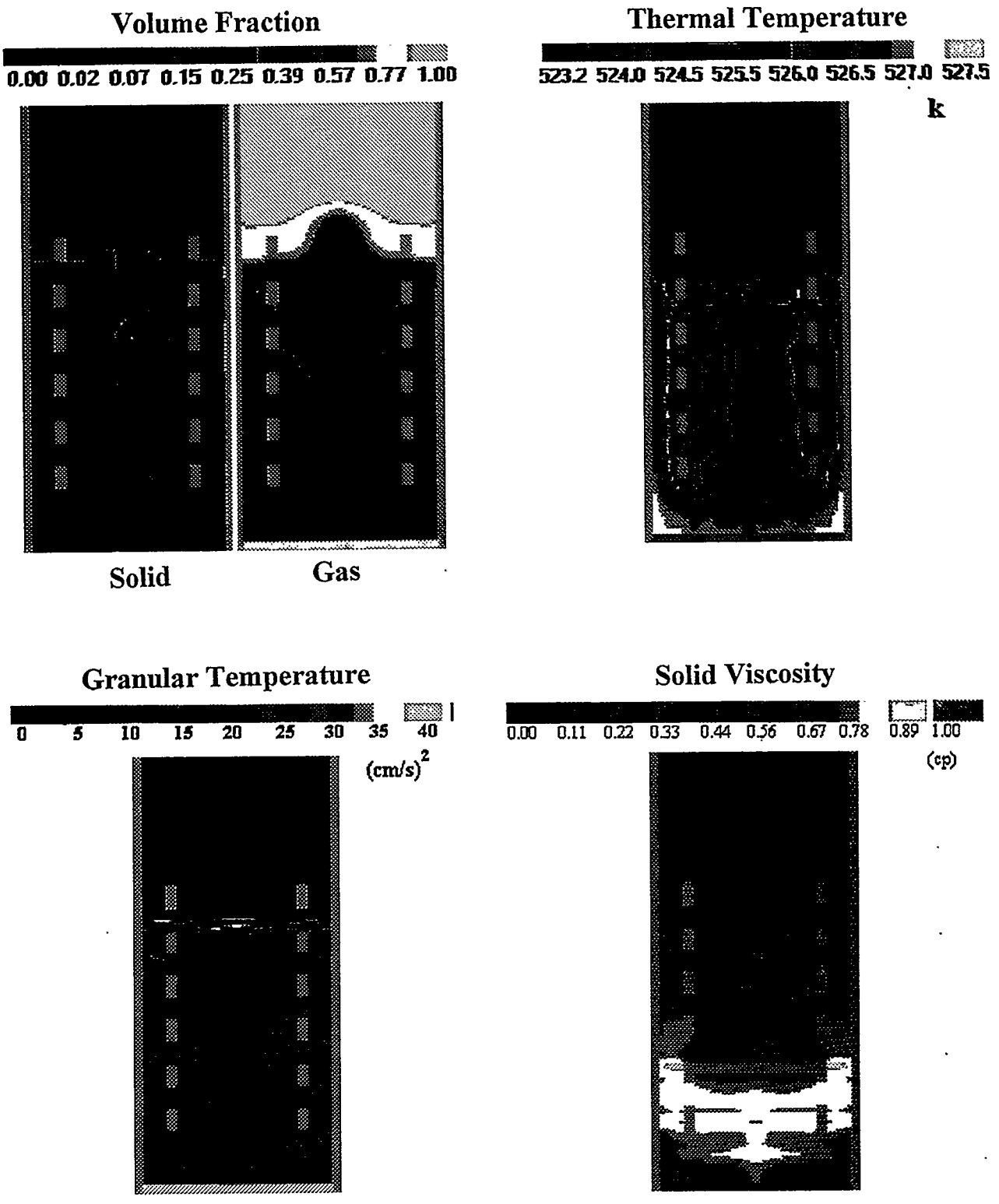

Figure 7 **Simulation of Air Products Slurry Bubble Column Reactor (RUN E8.1) at 12 seconds from Start-up.**

Figure 8 ROCKET MOTOR (1 ms after start-up)