# Modeling and simulation of multi-component, multi-phase fluid flows

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October 12, 2002

## Acknowledgments

The author would like to express sincere thanks and gratitude to his thesis advisor John Lowengrub for the encouragement and guidance.

The author is also thankful to Professor Ellen Longmire and Ilija N. Milosevic for an interesting joint project which is experimental and numerical program aimed at investigating topological transitions in liquid/liquid systems.

The author is thankful to Professor Chris Macosko and Jeffrey A. Galloway for an interesting another joint project investigating detection of cocontinuous structures in polymer blends.

The author gives thanks to Professor Vittorio Cristini for his useful comments,

Furthermore, the author is grateful to his colleagues at Minnesota for their help and assistance. In particular, thanks are due to Kyungkeun Kang and Jinhae Park for their joint-works with the author. Very special thanks are reserved for my brothers, sisters, and my wife Sumi Kim for her love.

Finally, the author would like to express his deepest gratitude toward mother for her infinite love and father for his life guidance.

# ABSTRACT

In this thesis, I study the dynamics of multi-component, multi-phase fluid flows that are present in a variety of industrial and scientific applications. To investigate the complex interactions that arise in such systems, I develop numerical algorithms and perform simulations. In particular, the methods that I have developed are capable of describing the evolution of deformable interfaces in a wide variety of physical situations including pinchoff and reconnection of interfaces.

I developed new finite-difference, multigrid numerical methods to solve the governing system of equations. This work includes the development of the non-linear multigrid methods to solve the Cahn-Hilliard equation. I also prove convergence of this new method.

In flows containing two liquid components, I investigated the pinchoff of a liquid jet. This is a fundamental problem for which much experimental and theoretical data has been obtained. My method recovers pinchoff due to the classical Rayleigh instability. In preliminary results, I approximate experimental inflow/outflow boundary conditions, I have obtained qualitative agreement with recent experiments by Professor Longmire and co-workers (U. Minn.) for both the jet shape and velocity and vorticity fields.

I also considered more complex two-phase flows. For example, I am investigating the development of co-continuous structures in polymer blends where a dispersed phase, under vigorous shear, undergoes a coalescence cascade and becomes continuous. Such systems have many important industrial applications. I have formulated an exact measure of cocontinuity and have performed three dimensional numerical simulations that show the cascade strongly depends on the volume fraction and weakly depends on the viscosity ratio of the fluids.

In flows containing three liquid components, I investigated immiscible compound drops and the effect of preferential mixing in the classical Rayleigh-Taylor instability. I found that a range of parameters exists for which a heavy drop may be made buoyant by encapsulation with lighter fluid and the resulting compound drop exists as a stable structure. In the case of preferential mixing, applicable to liquid-liquid extraction for example, I found that the instability significantly accelerates the mixing process.

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# Chapter 1

# Introduction

## 1.1 Physical Problems

Most engineering, industrial and biomedical applications of fluid flows involve fluids that contain more than one component. The components may be immiscible and thus separated by sharp interfaces, or they may display a partial miscibility such that one or more of the components may mix. Examples of the former include liquid/liquid extractors in which an emulsion of droplets of one fluid (e.g. water) are separated from a matrix fluid (e.g. oil) which is necessary for the transport of petroleum. Examples of the latter include remediation of a contaminated waste stream in which a contaminant is diffused across an interface from one immiscible fluid to another. The accurate description of the free surfaces in such flows is important and determines separation, mixing and reaction rates which thus control the efficiency of the process. For example, if one of the fluids is broken up into small droplets, the interfacial area and hence the mass transfer rates are increased. Topological transitions such as pinchoff and reconnection of interfaces are fundamental features of these flows and strongly affect the flow microstructure (localized morphology). A modeling and simulation effort must then accurately resolve these flow events.

The microstructure of multicomponent fluid flows also impacts their rheology. For example, emulsions containing monodisperse drops generally exhibit different macroscopic characteristics from those that contain a polydisperse droplet phase. In the case of polymer blends, the microstructure also offers a new and important route to materials with unique combinations of properties not available in a single polymer. For example, during processing of immiscible polymer blends, a number of non-equilibrium microstructures can form. By dropping the temperature of the blend, these microstructures can be frozen in the resulting solid yielding optimal material properties (e.g. stiffness, strength and toughness) under

certain conditions.

Problems involving multicomponent fluid flows where there are changes in interface topology are difficult to study theoretically and experimentally for several reasons. The fluids in which these transitions occur are complex with the density, viscosity, surface tension, and diffusivity, playing important roles in the transition process, affecting both the post-transition structure of the flow and the dynamics of the transition itself. The transitions typically result from a competition between flow instabilities (e.g. due to shear or density stratification), and stabilizing influences (e.g. due to surface tension and/or viscosity). A second problem associated with topological transitions is caused by the short time scales over which they occur.

Nevertheless, recent advances in experimental techniques by Professor Longmire and co-workers have made it possible, for the first time, to both visualize and to provide quantitative measurements of flows during pinchoff and reconnection events. One of the goals of this thesis is to use Professor Longmire's data to validate and refine a novel, physically-based modeling and simulation approach describing interfacial flows undergoing topological transitions. Theory and simulations can provide a unique insight to this phenomenon and can lead, for example, to determination of physically-based sharp interface reconnection conditions that can be used to develop accurate engineering-level models.

## 1.2 A Sharp Interface Description of Fluid Systems

In classical models, an interface between two immiscible fluids is treated as infinitely thin (sharp) across which densities, viscosities and the normal stress may have jumps. Fig. 1.1 illustrates jumps of these quantities across interface. The incompressible Navier-Stokes equations governing fluid flow in each flow component $i$ are

$$\begin{aligned} \nabla \cdot \mathbf{u} &= 0, \\ \rho_i(\mathbf{u}_t + \mathbf{u} \cdot \nabla \mathbf{u}) &= \nabla \cdot \mathbf{T} + \rho_i \mathbf{g}, \\ \mathbf{T} &= -p\mathbf{I} + \eta_i(\nabla \mathbf{u} + \nabla \mathbf{u}^T), \end{aligned} \tag{1.2.1}$$

where $\mathbf{u}$ is the fluid velocity field, $\rho_i$ is the constant fluid density of component $i$, $\mathbf{T}$ is the stress tensor, $p$ is the pressure field, $\mathbf{I}$ is the identity tensor, $\eta_i$ is fluid viscosity, and $\mathbf{g}$ is the gravitational force.

In these models, it is assumed that the interface has a surface tension, which on applying a stress balance at the interface gives rise to the interfacial boundary condition

$$\mathbf{T} \cdot \hat{n}|_{-}^{+} = \sigma \kappa \hat{n},$$

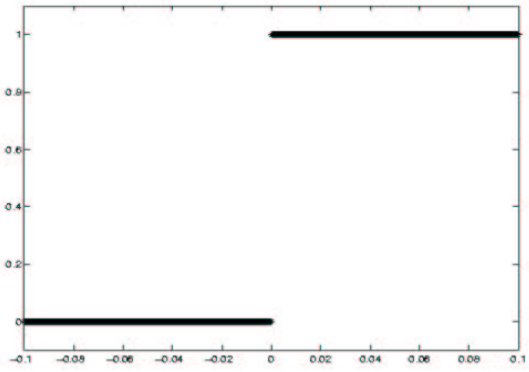

Figure 1.1: sharp interface

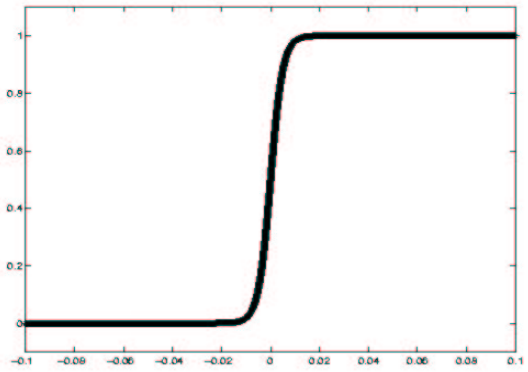

Figure 1.2: diffuse interface

which relates the jump (from inner to outer fluid) in the stress across the interface to the interfacial curvature. Here, $\hat{n}$ is the unit vector normal to the interface, $\sigma$ is the (constant) surface tension, and $\kappa$ is the total curvature ($\kappa = 2$ on a sphere of radius 1). Further, the interface is impermeable:

$$\mathbf{u} \cdot \hat{n}|_- = \mathbf{u} \cdot \hat{n}|_+ = V_n,$$

where $\mathbf{u}$ represents the velocity of the fluid and $V_n$ is the normal velocity of the interface.

Finally, for viscous fluids, the tangential velocity across the interface is also continuous:

$$[\mathbf{u} - (\mathbf{u} \cdot \hat{n})\hat{n}]|_-^+ = 0.$$

The above free-boundary description has been a highly successful simulation model when there are only a few interfaces that do not change topology during the flow evolution. Sharp interface-tracking methods explicitly treat the interface as a discontinuity. Usually, it is specified by an ordered set of marker points, connected by an interpolation surface. The markers are advected in a Lagrangian manner and then redistributed to obtain the best resolution of the interface [104]. Examples of tracking methods include boundary integral methods [13, 24, 25, 26, 33, 42] and finite-element variational methods [12, 134]. In the former approach, the dimensionality of the system is reduced by mapping the equations onto an interface surface. This can only be done in certain physical conditions such as in potential flow or in Stokes flow. The finite element approach has broader applicability since the equations are solved in the bulk as well as interface domains. However, in both cases the sharp interface description breaks-down when interfaces pinchoff or reconnect. This is reflected in development of singularities in the solutions of the governing equations. Such topological transitions are fundamental features of multi-component fluid flows.

Many researchers (see [94, 126] for example) have tried using *ad hoc* methods to change the topology of sharp interfaces. While this approach, often referred to as "contour surgery," allows topological transitions to be overcome, it is difficult to justify the reconnection conditions based on physical principles. Moreover, performing mesh surgery in three dimensions is very challenging. In a few special cases, all involving fluid-gas interfaces, it is possible to develop physically-based reconnection conditions by using special similarity solutions of the Navier-Stokes equations (see [86, 59, 60]). For flows involving liquid/liquid interfaces, however, where there is significant competition between inertia, viscosity and surface tension, the dynamics of topology transitions are much more complicated than in the fluid-gas case and no such similarity solutions are known.

## 1.3 A Distribution Description of Fluid Systems

Interface capturing methods provide an alternative approach to interface-tracking methods. In an interface capturing method such as the level set method (e.g. [41, 103, 112, 116, 117, 131, 132]), the interface is captured as the zero level set of a smooth function $\varphi(\mathbf{x}, t)$; i.e., the interface $\Gamma(t) = \{\mathbf{x}|\varphi(\mathbf{x}, t) = 0\}$. $\varphi$ is positive inside region $\Omega$, negative outside $\Omega$, and is zero on $\Gamma(t)$. The $\varphi$ values (levels) are convected with the velocity field $\mathbf{u}$:

$$\frac{\partial \varphi}{\partial t} + \mathbf{u} \cdot \nabla \varphi = 0.$$

The Navier-Stokes equations and boundary conditions can then be written in distribution form as

$$\begin{aligned} \nabla \cdot \mathbf{u} &= 0, \\ \rho(\varphi)(\mathbf{u}_t + \mathbf{u} \cdot \nabla \mathbf{u}) &= \nabla \cdot \mathbf{T} - \kappa \delta(\varphi) \hat{\mathbf{n}} + \rho \mathbf{g}, \\ \mathbf{T} &= -p\mathbf{I} + \eta(\nabla \mathbf{u} + \nabla \mathbf{u}^T), \end{aligned} \tag{1.3.2}$$

where $\hat{\mathbf{n}} = \dfrac{\nabla \varphi}{|\nabla \varphi|}$ is the unit normal to $\Gamma$, $\kappa = \nabla \cdot \hat{\mathbf{n}}$ is the total curvature of the interface and $\delta(\varphi)$ is the one dimensional delta function.

The different level sets (or fixed values of the function $\varphi$) can exhibit different topologies. Typically, the interface is effectively given a finite thickness by smoothing the flow discontinuities (e.g. density, viscosity) over a narrow region surrounding $\Gamma$. Although this procedure generally yields a smooth evolution through topological changes, one can demonstrate that the results can depend essentially on the type of smoothing chosen [93]. Within the context of the level set method, it is not clear which types of smoothing are physically justified.

In an alternative approach, the volume of fluid (VOF) method (e.g. [75, 119]) uses a volume fraction distribution to represent the two fluid phases. The singular surface force is evaluated from the volume

fraction in a way analogous to that described above for the level set approach. The fields for all variables and properties are shared by the phases and represent volume-averaged values, as long as the volume fraction of each of the phases is known at each location. Thus the variables and properties in any given cell are either purely representative of one of the phases or representative of a mixture of the phases, depending upon the volume fraction values. Based on the local value of the volume fraction, the appropriate properties and variables are assigned to each control volume within the domain. In the VOF approach, capturing topology changes requires the use of mathematical and numerical smoothing techniques (e.g. upwind methods). Again, it is not clear which types of smoothing are physically justified.

In recent work, Lowengrub and Truskinovsky [93] proposed a regularization of the sharp interface approach in which limited mixing between the fluids is allowed to occur across the interfacial zone ('partial miscibility regularization', PMR). From physical chemistry, it is well known that limited molecular mixing (or chemical diffusion) occurs between macroscopically "immiscible" fluids. This limited mixing provides a *physical mechanism* to smooth the flow discontinuities and to yield smooth evolutions through topological changes. Determining the accuracy and versatility of this model is a primary goal of this thesis.

## 1.4 A Diffuse-Interface Description of Fluid Systems

The diffuse interface method considered here was derived by Lowengrub & Truskinovsky [93] and is a physically-based model to compute flows with changes in interface topology. The model, which incorporates buoyancy, viscosity, compressibility, surface tension, and chemical diffusivity at interfaces, allows topological transitions to occur without relying on ad hoc 'cut and connect' or smoothing procedures. The diffusion is limited if the components are macroscopically immiscible and reflects the partial miscibility real fluids always display. As a consequence, interfaces separating the flow components are diffuse although the thickness of interfaces between real immiscible fluids, such as oil and water, may be nanoscopic in many circumstances (see the Fig. 1.2). The interface thickness (denoted by $\epsilon > 0$ below) and structure may be important at topological transitions since the distance between interfaces becomes comparable to the interface thickness. It is interesting to note that the idea that interfaces have a finite thickness dates to Poisson [106] and Gibbs [64]. The idea is this: according to the thermodynamics of immiscible fluids, there is a range of concentrations (of one of the components) where the free energy is concave and homogeneous states are unstable [64]. An interface between two immiscible fluids may then be described as a layer where thermodynamically unstable mixtures are stabilized by weakly non-local terms (gradients) in the energy [128] which have their origins in molecular force interactions between the

components [109, 50]. These gradient terms induce extra reactive stresses in the fluid which become surface stress in the zero thickness limit. In this diffuse-interface model, concentration fields are introduced and are coupled to the fluid motion. The resulting system involves the coupling of the Navier-Stokes (NS) equations, with extra reactive stress $-\frac{\sigma}{\epsilon}\rho c\nabla\mu$ due to concentration gradients that mimics the force due to the surface tension $\sigma$, to a nonlinear, fourth order diffusion equation of Cahn-Hilliard (CH, [31]) type for the concentration. The Navier-Stokes system is

$$\begin{aligned} \nabla\cdot\mathbf{u} &= \alpha\nabla\cdot(M(c)\nabla\mu)\,, \\ \rho(c)(\mathbf{u}_t+\mathbf{u}\cdot\nabla\mathbf{u}) &= \rho\nabla\tilde{p}-\frac{\sigma}{\epsilon}\rho c\nabla\mu+\nabla\cdot\left(\eta(c)(\nabla\mathbf{u}+\nabla\mathbf{u}^T)\right)+\rho\mathbf{g}, \end{aligned} \tag{1.4.3}$$

where $c(\mathbf{x},t)$ is the mass ratio of one of the components in a heterogeneous mixture of two fluids (e.g. $m_1/m$ where $m_1$ is the mass of component 1 and $m$ is the total mass of the binary fluid in a representative volume $V$) and $\alpha=-\rho'/\rho^2$ and $\rho'=\dfrac{d\rho}{dc}$ and $1/\rho(c)=c/\rho_1+(1-c)/\rho_2$ is the density ($\rho_1$ and $\rho_2$ are the constant densities of the components). The chemical potential $\mu$ is given by

$$\mu=F'(c)-\alpha\epsilon\tilde{p}-\epsilon^2\Delta c$$

where $F(c)$ is the Helmholtz free energy and $\tilde{p}$ is the modified pressure:

$$\tilde{p}=\frac{p}{\rho}+\frac{\rho\epsilon}{2}|\nabla c|^2+\frac{1}{\epsilon}F(c).$$

The Cahn-Hilliard equation is

$$\rho(c_t+\mathbf{u}\cdot\nabla c)=\nabla\cdot(M(c)\nabla\mu), \tag{1.4.4}$$

where $M(c)$ is a degenerate mobility often taken to be $M(c)=c(1-c)$.

This system is capable of describing flows with both miscible and immiscible components. If $F(c)$ is convex, the components are miscible. If $F(c)$ is non-convex, the components are immiscible on a macroscopic scale. The system (1.4.3)-(1.4.4) has an associated non-increasing energy functional $\int_\Omega(\rho|\mathbf{u}|^2/2+\sigma F(c)/\epsilon+\sigma\epsilon|\nabla c|^2/2)\,d\Omega$. Moreover, Liu and Shkoller (preprint, 2001) recently proved rigorously that this system converges to the classical sharp interface model as $\epsilon\to 0$.

There has been much recent work on the use of diffuse interfaces in multi-component fluid flows and we refer the interested reader to the review of recent research by Anderson, McFadden & Wheeler [4]. To our knowledge, the study presented here are the first direct comparisons of these diffuse interface methods to experiments. Further, the numerical methods developed in this thesis are more efficient and accurate than other methods that currently appear in the literature.

The main achievements of this thesis are as follows:

- The development of new finite-difference, multigrid numerical methods to solve the governing system of equations. This work includes the development of the non-linear multigrid methods to solve the Cahn-Hilliard (fourth order advection-diffusion) equation [31]. Convergence of this new method is proved.

- The investigation of the pinchoff of a liquid jet and comparison to experiments. This is a fundamental problem for which much experimental and theoretical data has been obtained. My method recovers pinchoff due to the classical Rayleigh instability. In preliminary results, in which I approximate experimental inflow/outflow boundary conditions, I have obtained qualitative agreement with recent experiments by Professor Longmire and co-workers (U. Minn.) for both the jet shape and velocity and vorticity fields.

- The investigation of the development of co-continuous structures in polymer blends where a dispersed phase, under vigorous shear, undergoes a coalescence cascade and becomes continuous. Such systems have many important industrial applications. I have formulated an exact measure of cocontinuity and have performed three dimensional numerical simulations that show the cascade strongly depends on the volume fraction and weakly depends on the viscosity ratio of the fluids.

- The investigation of immiscible compound drops and the effect of preferential mixing in the classical Rayleigh-Taylor instability. I found that range of parameters exists in which a heavy drop may be made buoyant by encapsulation with lighter fluid and the resulting compound drop exists as a stable structure. In the case of preferential mixing, applicable to liquid-liquid extraction for example, I found that the instability significantly accelerates the mixing process.