Efficient Modeling of Multicomponent Diffusive Mixing

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Motivation: Giant Fluctuations

• Experiment: mixing of water and various denser organic compounds ((a) urea, (b) glycerol, (c) polyethylene, (d) lysozyme) with initially flat interface

• Shadowgraph techniques reveal giant fluctuations caused by thermal fluctuations develop over several minutes.

Not an instability!

Motivation: Mixed-Mode Instability

- Experiment: ternary mixture (water, salt, sugar) between two glass plates.
  - Heavy saltwater on top of light sugar water
  - Salt diffuses into water 4x faster than sugar

Carballido-Landeira et al., Physics of Fluids, 2013
We are interested in developing a model for multicomponent diffusive mixing at length scales where thermal fluctuations are important.

- Arbitrary number of fluid components, non-ideal space/time-dependent transport properties, thermal fluctuations.
- Large Schmidt numbers (momentum diffusion divided by mass diffusion) requires implicit treatment of viscosity.
- Requires long-time integration (minutes to hours). More established particle or compressible methods are not fast enough.
Low Mach Number Fluctuating Hydrodynamics

• We develop a Low Mach number continuum model.
  – Begin with the compressible equations of fluctuating hydrodynamics (Landau & Lifschitz).
  – Make the assumption that acoustic waves are unimportant to the overall solution.
  – Using low Mach number asymptotics, we derive an equation set that mathematically eliminates sound waves and enforces instantaneous acoustic equilibration.
  – Obtain a divergence constraint on velocity (similar to incompressible Navier Stokes) but the divergence is determined by the mixing of fluids.
  – Model allows for larger advective-based time steps.
Low Mach Number Fluctuating Hydrodynamics

- Given an arbitrary number of fluid components with pure densities, $\bar{\rho}_i$
- The total density is the sum of the component densities, $\rho = \sum \rho_i$
- We enforce no volume change upon mixing, $\sum \frac{\rho_i}{\bar{\rho}_i} = 1$
- The resulting low Mach number model is

$$\frac{\partial (\rho \mathbf{v})}{\partial t} = - \nabla \cdot (\rho \mathbf{v} \mathbf{v}^T) - \nabla \pi + \nabla \cdot \mathbf{\tau} (\mathbf{v}) + \nabla \cdot \mathbf{\Sigma} + \rho \mathbf{g}$$

$$\frac{\partial \rho_i}{\partial t} = - \nabla \cdot (\rho_i \mathbf{v}) + \nabla \cdot \mathbf{F}_i$$

Divergence constraint represents “no volume change upon mixing”

Model for stochastic forcing (Landau, Lifschitz)

Multicomponent non-ideal diffusion (based on works by Kuiken, Giovangigli, Kjelstrup) and stochastic forcing (Ottinger)
Numerical Features

- Staggered grid velocity finite volume formulation
- Arbitrary number of fluid components
- Non-ideal multicomponent diffusion with space/time-varying transport coefficients
- Stokes solver for coupled viscous/projection problem with no loss of accuracy at boundaries
- Projection method preconditioner for Stokes solver; we have demonstrated the overall algorithm is competitive with standard operator split approaches
- Multiple time stepping schemes to support inertial and large Schmidt number (via Stokes approximation) regimes
- Multistage centered scalar advection for stochastic flow; option for higher-order Godunov schemes for deterministic flow
- Implemented in highly scalable BoxLib software framework developed at Berkeley Lab.
Model Validation

- Diffusive mixing of two fluids with density contrast 4.
  - Left: Molecular dynamics simulation (Skoge 2006, publicly available)
  - Right: Low Mach number fluctuating hydrodynamics
Analysis of Interface Fluctuations

- Discrete spatial spectrum of interface fluctuations shows excellent agreement.
Giant Fluctuations

- We are able to reproduce giant fluctuations observed in stable fluid configurations

Mixed Mode Instability

• Simulation parameters match experimental setup for ternary mixed-mode instability.

Carballido-Landeira et al., Physics of Fluids, 2013
References

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• A. Donev et al., "Low Mach Number Fluctuating Hydrodynamics of Multispecies Liquid Mixtures", *in preparation*