SAND2011-5896C

Modeling Polyurethane Foam Expansion Using A Finite Element/Level Set Method

Rekha Rao, Lisa Mondy, David Noble, Doug Adolf, Mat Celina, Ed Russick,
Jamie Kropka, and Ray Cote
Sandia National Laboratories
Albuquerque, NM

James Mahoney
Honeywell Kansas City Plant
Kansas City, MO

11th Nuclear Weapons Engineering Analysis Conference
Kirtland Air Force Base, NM
September 6th -8th, 2011

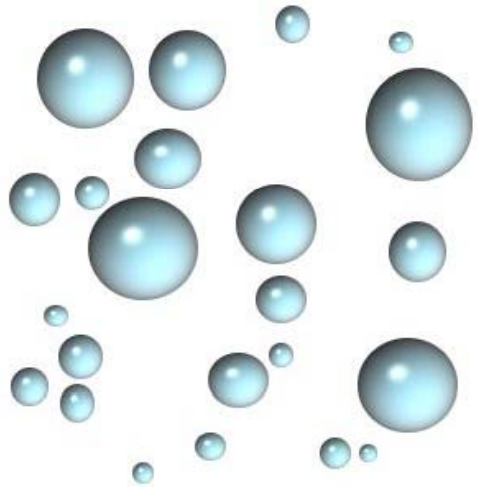
Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

National Nuclear Security Administration

LDRD
LABORATORY DIRECTED RESEARCH & DEVELOPMENT

Sandia National Laboratories

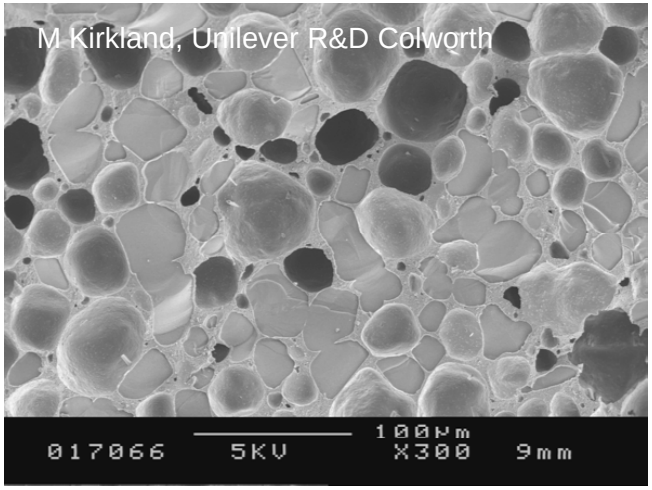
What is a Foam?



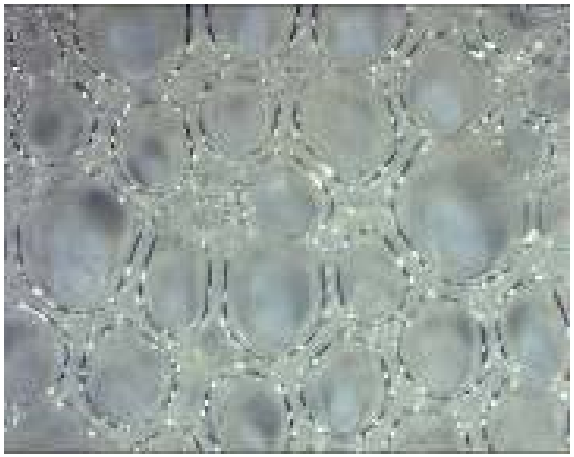
Bubbles

Whipped cream

- A multiphase material of gas bubbles in a liquid or solid matrix
- How do you make a foam?
  - Generate bubbles in a liquid
  - Stabilize them with particles, fat globules, or surfactant
  - Solidify liquid -freezing, polymerization, or phase change – if desired



Ice cream is a foam – that's why it is so much work to make



Epoxy foam is a collection of bubbles in polymer

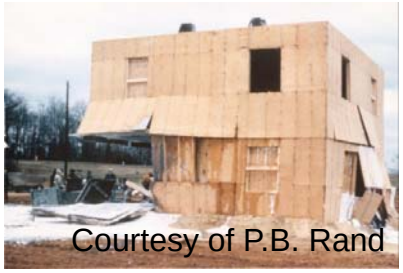
Foams need enough bubbles to jam, *e.g.* bubbles are touching or it is just a bubbly liquid

Sandia's interest in foam

Explosion Suppression

no foam

foam



Courtesy of P.B. Rand

Aubert et al. *Scientific American* 254 74 (1986)

Decontamination

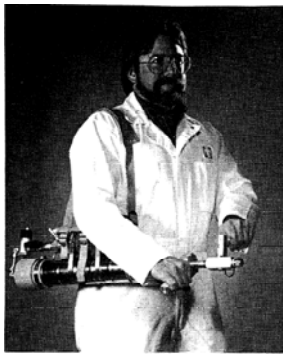
Courtesy of J.B. Kelley

Structural

Courtesy of R. Montoya; Whinnery SAND2006-6855C

Encapsulation

Intruders/Unruly Crowds

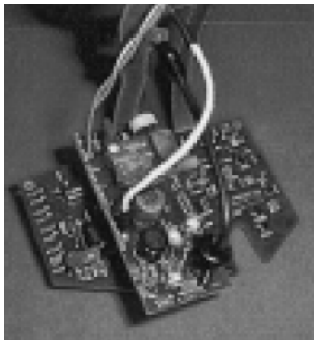


Scott *SAND096-2495C*; Russick *SAND2002-1103P*

Electronics—removable foam

reversible chemistry

90°C



McElhanon et al. *J. Appl. Polymer Sci.* 85 1496 (2002)

Sandia National Laboratories

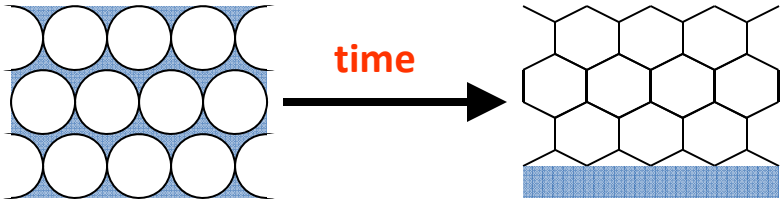
Liquid foam characterization is challenging

Opacity prevents direct observation

- Foams are multiphase materials with a compressible gas dispersed as bubbles in a continuous phase
- Bubble microstructure affects macroscopic properties
- Microstructure can evolve in reversible and irreversible manner
- Property measurements can alter foam

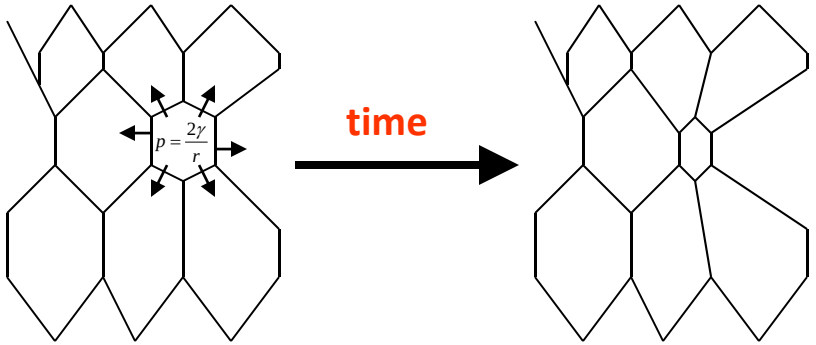
Structure is continuously evolving

Liquid Drainage



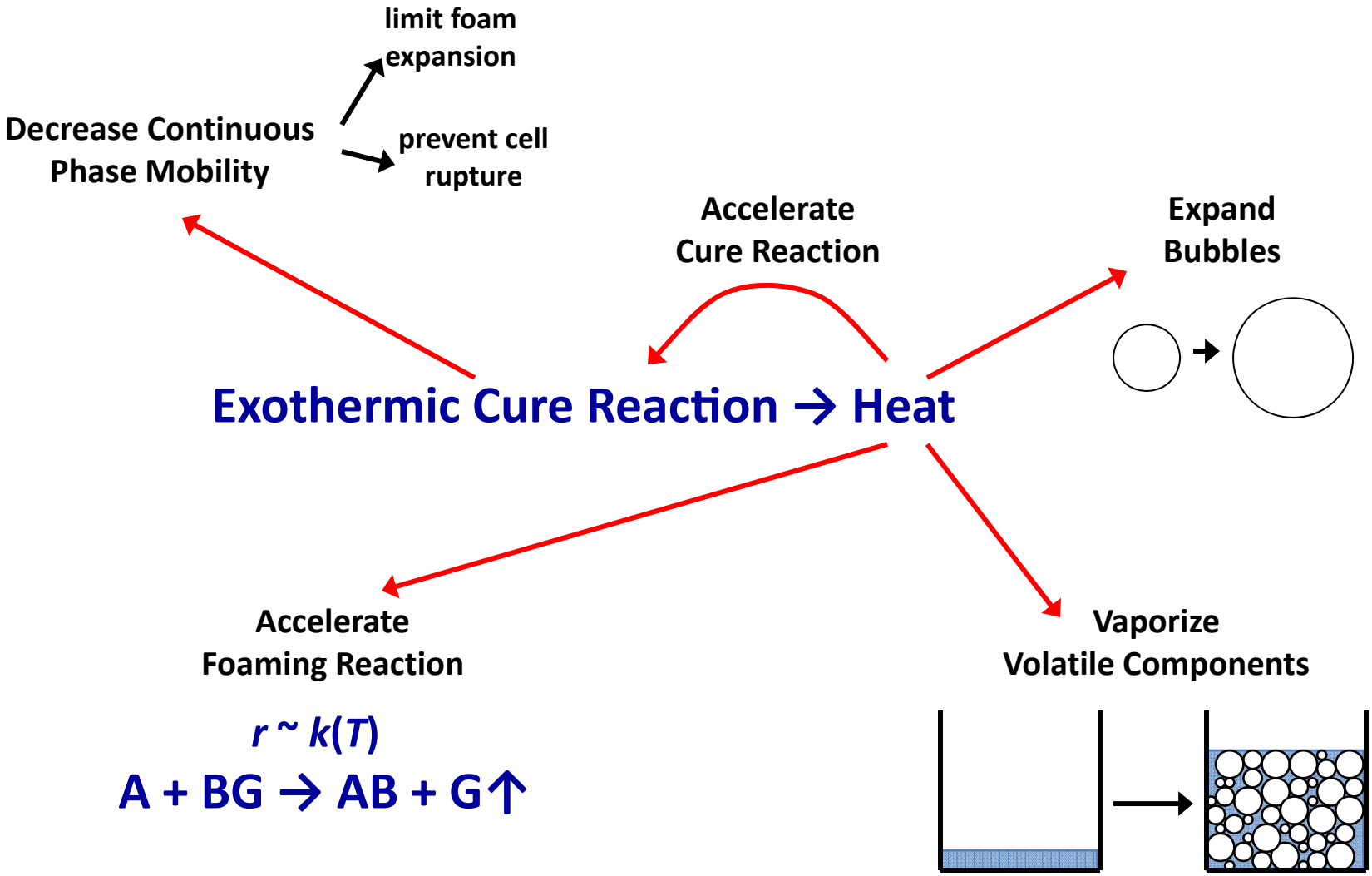
Coalescence and rupture also occur

Cell Coarsening



Sandia National Laboratories

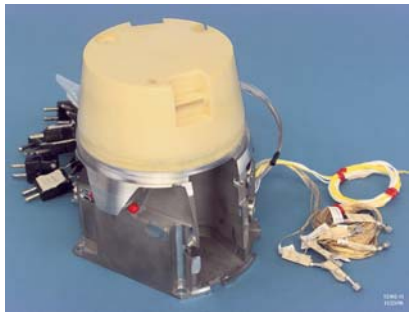
Polymeric foams offer additional complexities and are difficult to understand on a fundamental basis



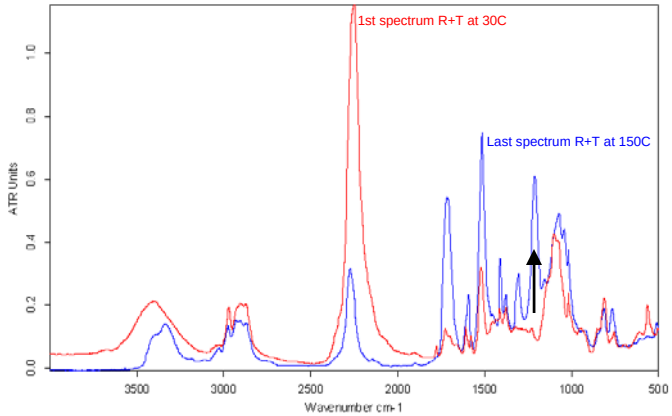
Sandia National Laboratories

Polyurethane (PMDI-4): Model Development

- At Sandia, we use a variety of physically and chemically blown foams.
- PMDI-4 is used as an encapsulant for electronic components, to mitigate against shock and vibration
- We would like to develop a computational model to help us understand foam expansion for manufacturing applications.
- Polyurethane is a chemically blown foam: fundamentally different model is needed from physically blown foams.
- Polyurethane foams have two primary, competing simultaneous reactions: CO_2 production and polymerization. Separating these reactions can be difficult.
- DSC does not offer enough resolution: Used IR to track reaction rates in several isothermal experiments at different temperatures.
- IR does not provide a clear signal for the foaming reaction: Gas generation measured by free rise height as before.



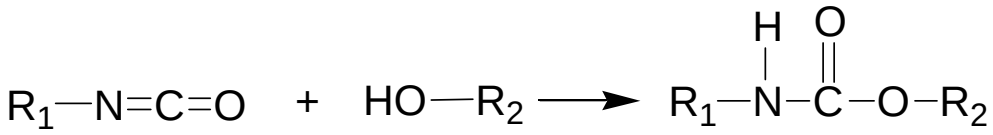
Component encapsulated with PMDI from "KCP Encapsulation Design Guide" (Mike Gerding)



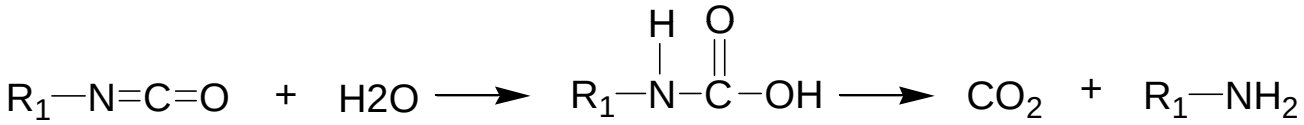
Peak 1218 represents pure curing reactions: polyol-isocyanate urethane reactions

Polyurethane Resin Cure and Foaming Reactions

Two key reactions: **Isocyanate reaction with polyols and water**

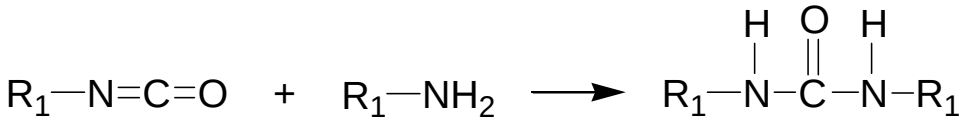


Urethane formation, crosslinking

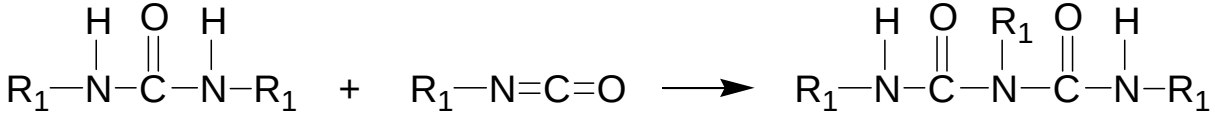


Foaming reaction yields CO_2 and amine

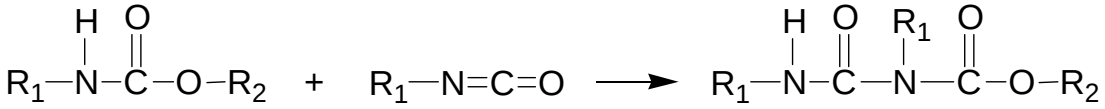
Various follow up reactions: **Isocyanate reaction with amine, urea and urethane**



Urea formation

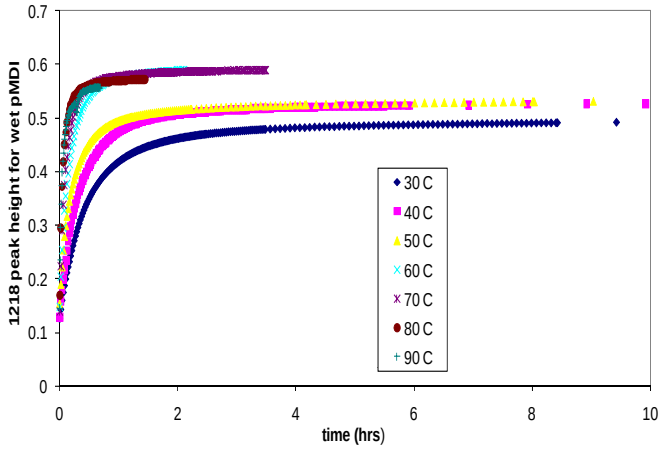


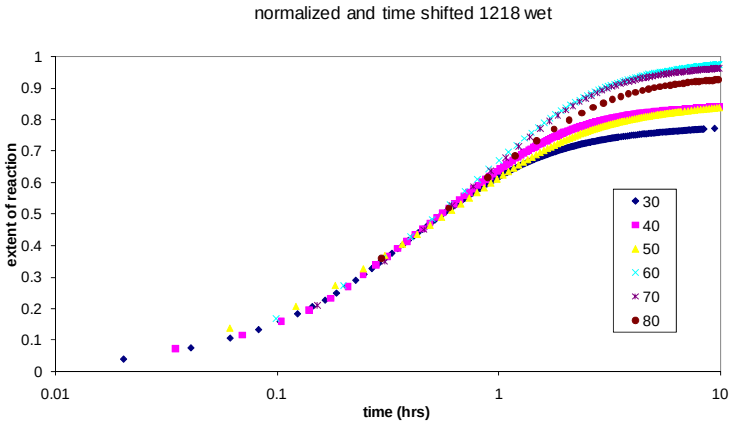
Biuret formation



Allophanate formation

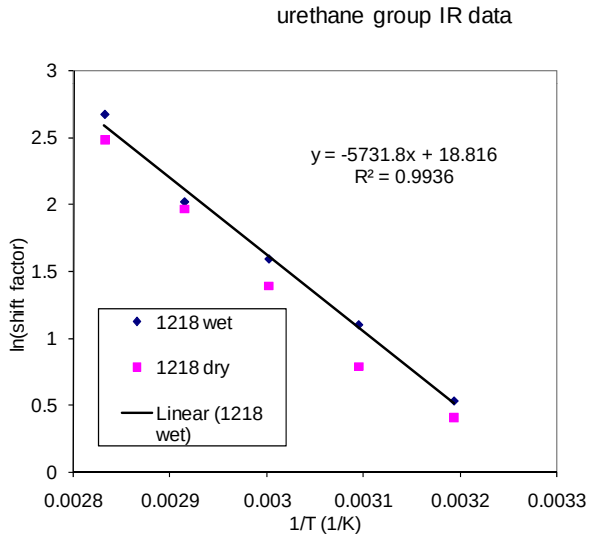
Extent of Reaction for Polymerization





- Shifted extent of reaction for isothermal tests carried out for various temperatures ranging from 30°C to 80°C.

- Peak height as a function of time for the 1218 cm^{-1} peak and the wet PMDI material. Isothermal tests were carried out for various temperatures ranging from 30°C to 90°C.



- Natural log of the shift factor versus the reciprocal temperature in Kelvin, gives the activation energy for the Arrhenius rate constant for the polymerization reaction.

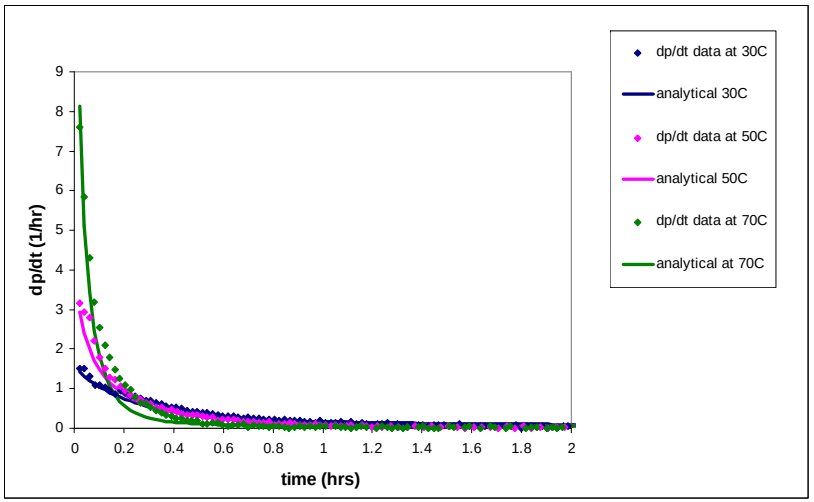
Extent of Reaction for Polymerization

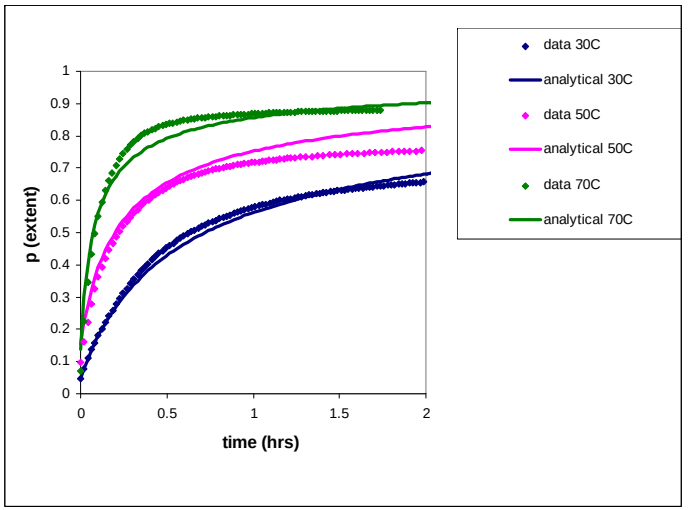
- Normalize the peak height data by the maximum height at the highest temperature to obtain the extent of reaction, p
- Superposition of data from different temperatures T gives activation energy ΔE
- Numerically differentiate the extent of reaction to obtain the rate
- Fit the rate and the extent of reaction simultaneously to a standard equation form, where only the exponent is unknown
- Form of between 2nd and 3rd order reaction fits data

$$\frac{d\xi_{cure}}{dt} = k_0 e^{\Delta E/RT}\left(1-\xi_{cure}\right)^{2.75}$$

k_0=2.96 x 10^8 1/hr,
ΔE/R=-5731.8 K

- "Wet" vs. "dry" slightly different rates – used full PMDI-4 (wet) formulation results

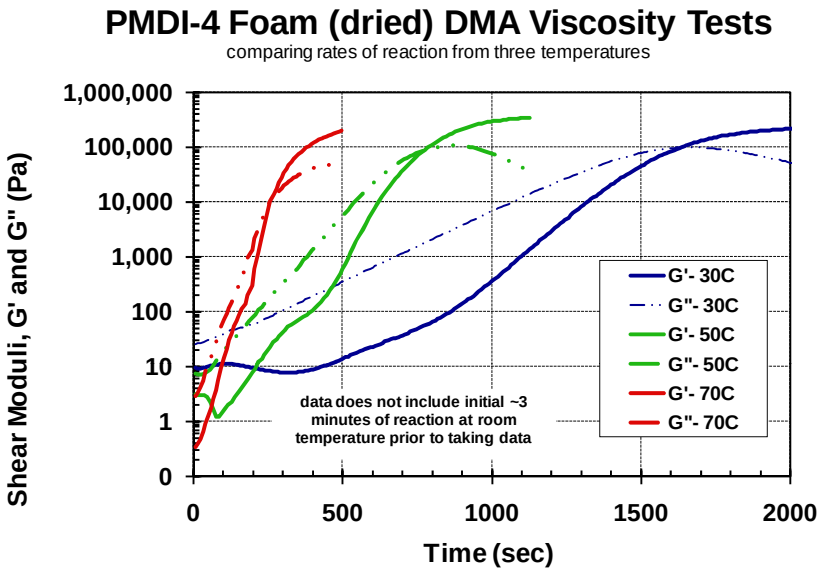


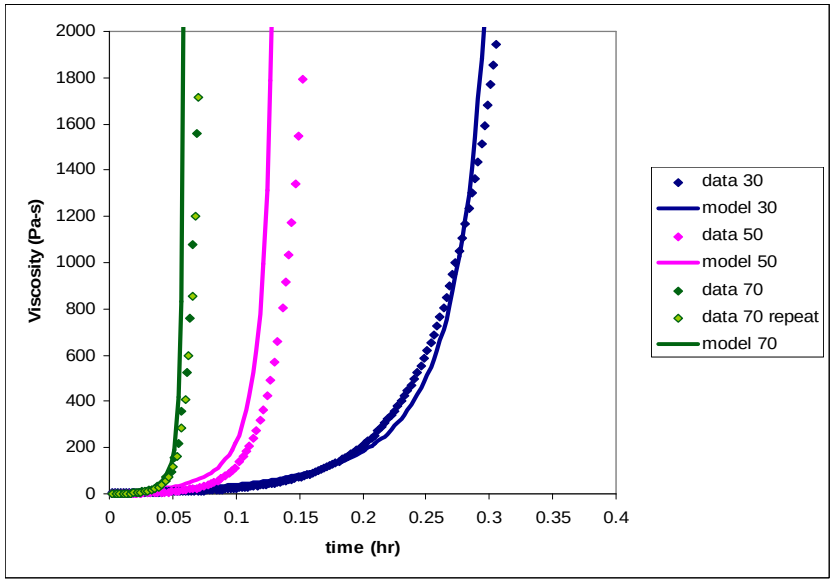


From polyol-isocyanate urethane reactions (Peak 1218 in PMDI-4 foaming)

Resin Continuous Phase Viscosity

- Storage and loss modulus for dry polyurethane at 30°C, 50°C, and 70°C measure in oscillatory rheometer
- The cross over point of G' and G'' gives the gel point and gel time of the polymer (0.46).
- Viscosity is correlated to extent of reaction and compared to data

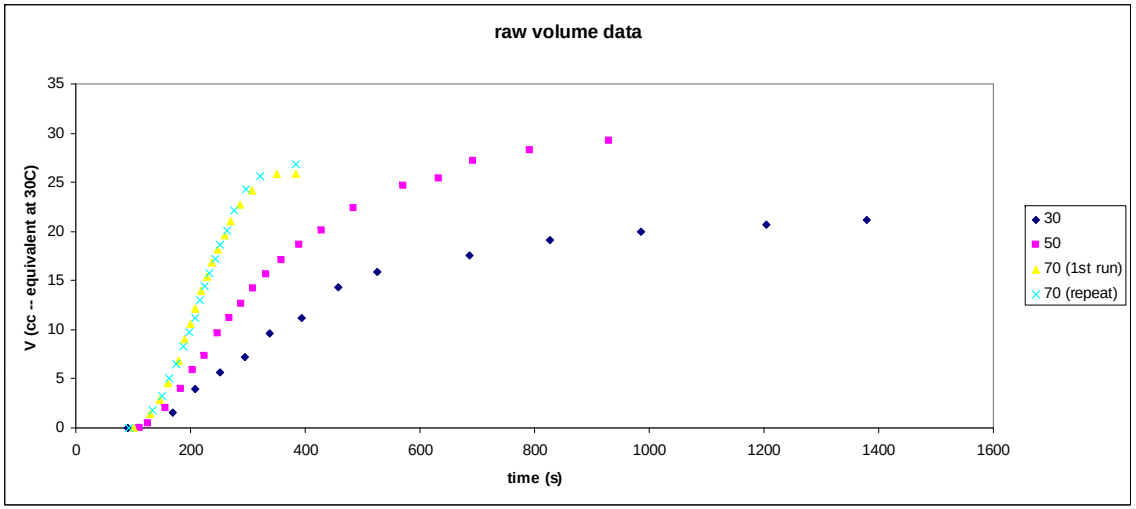




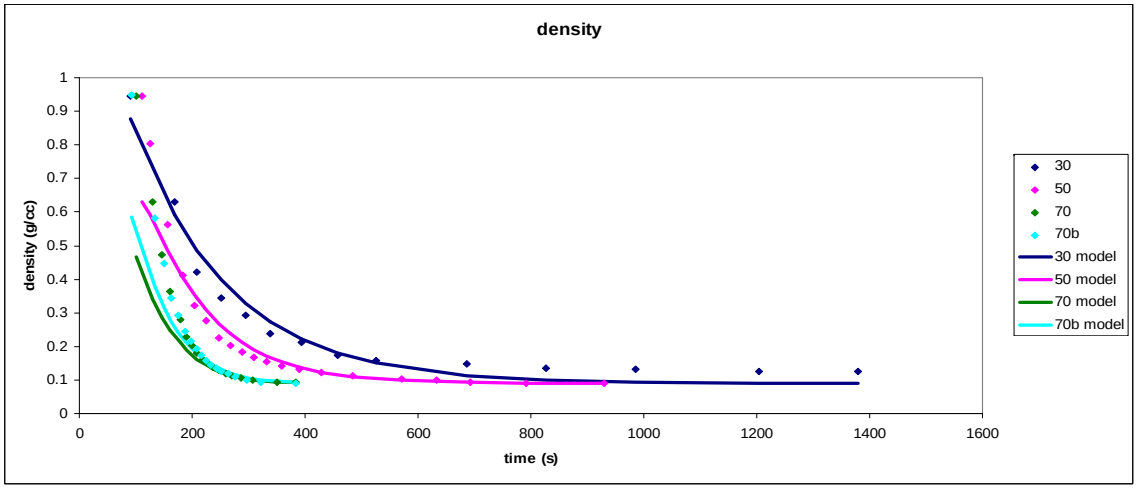
$$\eta_{cure} = \eta_0^0 \left(\frac{\xi_c - \xi}{\xi_c}\right)^{-2.0}, \xi_c = f(T)$$

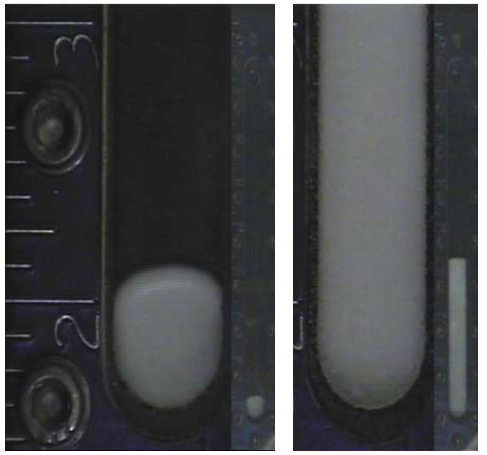
$$\eta_0^0 = 2.7x10^{-9} \exp\left(\frac{6.4kcal/(molK)}{RT}\right) Poise$$

Polyurethane: CO_2 Generation



$$\rho = (\rho_i - \rho_f)\exp\left[\frac{-(t-t_i)}{C(T)}\right] + \rho_f$$





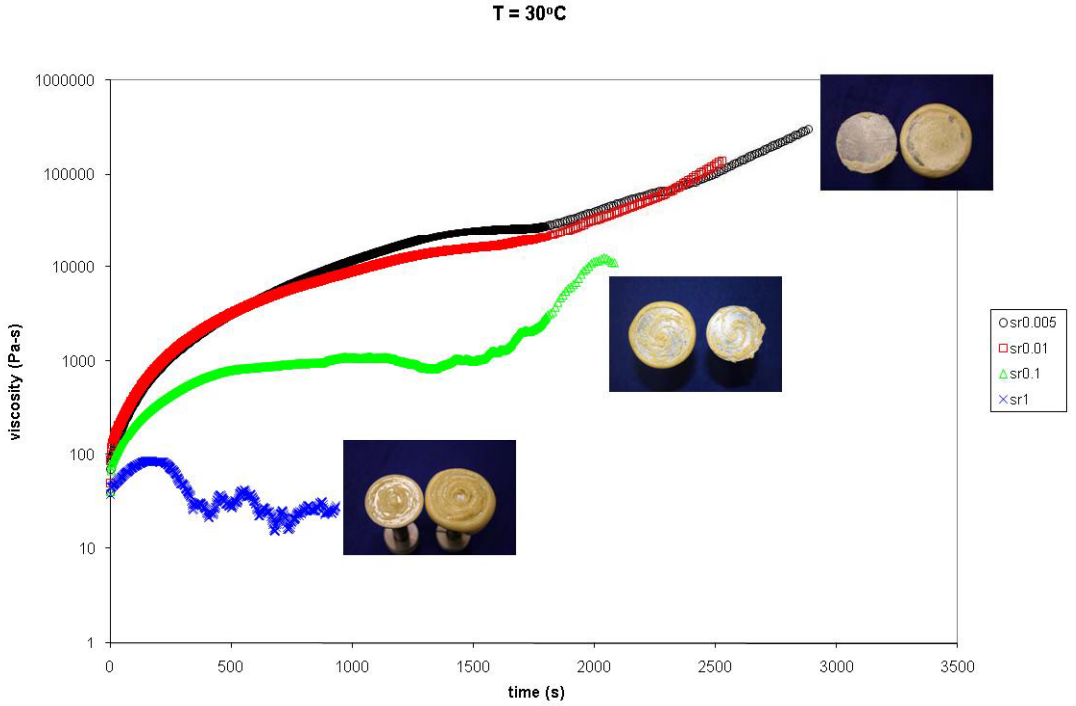
- 1st generation model uses a source term for gas generation
- Data from height of foam in a column at three temperatures (volume vs. time)
- Assume all gas is CO_2 generated in the reaction
- Foaming stops because curing continuous phase reaches gel point or resin become too viscoelastic to allow bubbles to expand

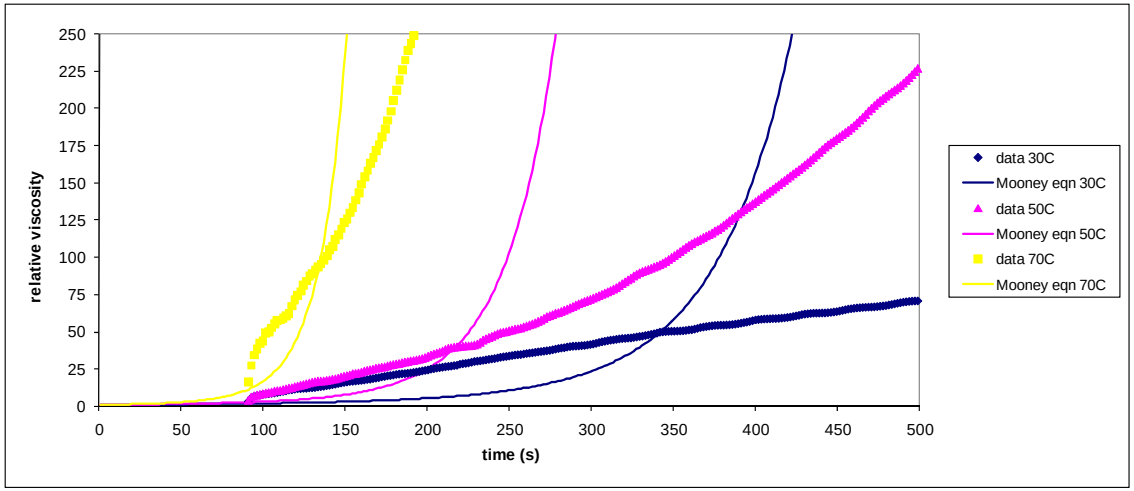
Polyurethane: Foam Viscosity

- Foam viscosity measured at 30°C, 50°C, and 70°C in oscillatory rheometer at various shear rates
- Relative viscosity as a function of time and temperature from rheological measurements on PMDI-4
- Taylor-Mooney Correlation gives the gas volume fraction dependence of the foam

$$\eta_{foam} = \eta_{cure} \exp\left(\frac{\phi_{gas}}{1-\phi_{gas}}\right)$$

- Correlation does fairly well at 70°C, but over predicts viscosity at lower temperatures





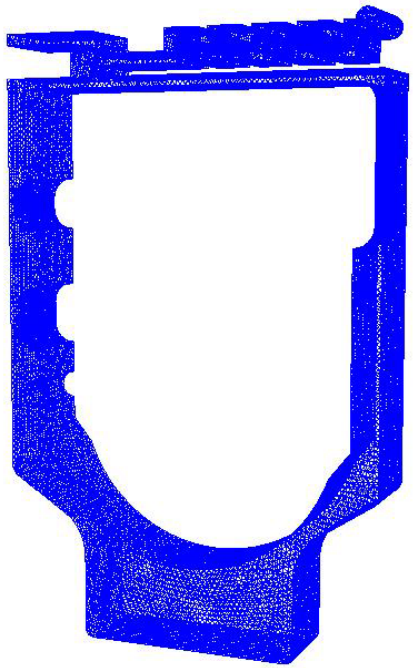
Higher Fidelity Model Adds More Complex Material Models with Cure, Temperature, and Void Fraction Dependence

$$\rho\frac{\partial \mathbf{v}}{\partial t} = -\rho\mathbf{v}\bullet\nabla\mathbf{v} - \nabla p + \nabla\bullet(\mu_f(\nabla\mathbf{v} + \nabla\mathbf{v}^t)) - \nabla\bullet\lambda(\nabla\bullet\mathbf{v})I + \rho\mathbf{g}$$

$$\rho C_{pf}\frac{\partial T}{\partial t} + \rho C_{pf} v\bullet\nabla T = \nabla\bullet(k\nabla T) + \rho\phi_e\Delta H_{rxn}\frac{\partial\xi}{\partial t} - \rho\lambda_{evap}\frac{\partial\phi_l}{\partial t}$$

$$\nabla\bullet\mathbf{v} = -\frac{1}{\rho_f}\left(\frac{\partial\rho_f}{\partial t} + \mathbf{v}\bullet\nabla\rho_f\right)$$

$$\frac{\partial\xi}{\partial t} + \nabla\bullet(\xi v) = k^i e^{\Delta E/RT}(1-\xi)^n$$



$$q = h(T - T_{oven})$$

$$n\bullet\tau\bullet t = \frac{1}{\beta}(v - v_s)\bullet t$$

$$v\bullet n = 0$$

$$\rho = \rho_{final} + (\rho_{initial} - \rho_{final})e^{-at} \qquad \text{a=f(T)}$$

$$\mu = \mu_0\exp\left(\frac{\varphi_v}{1-\varphi_v}\right) \qquad \mu_0 = \mu_0^0\exp\left(\frac{E_\mu}{RT}\right)\left(\frac{\xi_c^{\,2} - \xi^2}{\xi_c^2}\right)^{-4/3}$$

$$\lambda = \frac{4}{3}\mu_0\frac{(\phi_v - 1)}{\phi_v}$$

$$k = \frac{2}{3}\left(\frac{\rho}{\rho_e}\right)k_e + \left(1 - \frac{\rho}{\rho_e}\right)k_v$$

$$C_{pf} = C_{pl}\phi_l + C_{pv}\phi_v + C_{pe}\phi_e$$

Time scale and filling behavior set by density equation and unaffected by increasing shear viscosity or dilatational effects => Results here using constant viscosity (Rao et al, 2008)

Finite Element Implementation

- **Approximate variables with trial function, e.g.**

$$u \approx \sum_{i=1}^{n} u_i N_i \quad v \approx \sum_{i=1}^{n} v_i N_i \quad w \approx \sum_{i=1}^{n} w_i N_i \quad p \approx \sum_{i=1}^{m} p_i N_i^{'}$$

- **Substitute into equations of motion, weight residual with shape function for Galerkin implementation**

$$Weighted\text{-}Residual = \int N_i R_i dV$$

- **Gaussian quadrature**
- **Solve discretized system**

$$\underline{\underline{A}}\,\underline{x} = \underline{b}$$

- **Issues: Linear system solved with Krylov-Based iterative solvers => require stabilization**

Sandia National Laboratories

Evolving Level Set ϕ to Locate Foam-Gas Interface

•Given fluid velocity field, u(x,y,z), evolution on a fixed mesh is according to:

$$\frac{\partial \phi}{\partial t} + u \cdot \nabla \phi = 0$$

•Purely hyperbolic equation … fluid particles on $\phi(x,y,z) = 0$ should stay on this contour indefinitely

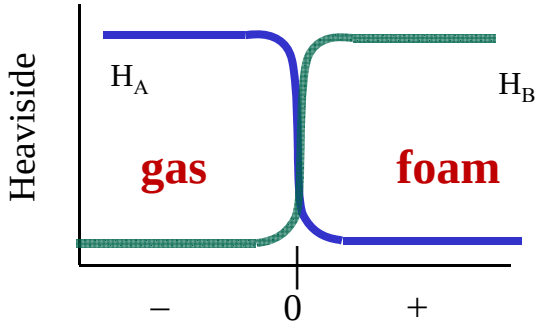
- Does not preserve $\phi(x,y,z)$ as a distance function
- Introduces renormalization step.

•Equations are averaged depending on the level set, ϕ

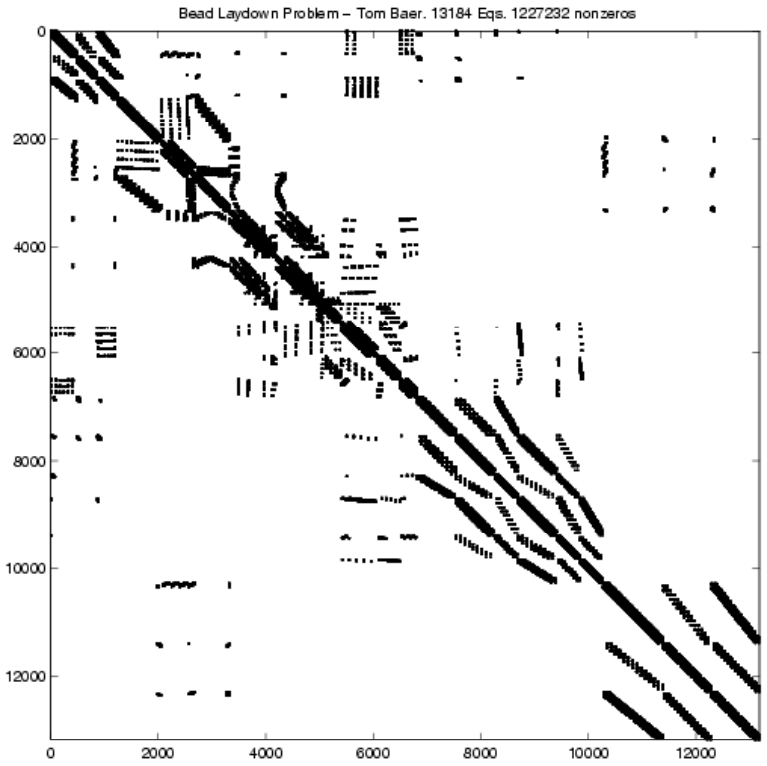
$$H_A \rho_A \frac{Du}{Dt} + H_B \rho_B \frac{Du}{Dt} = -\nabla P + H_A \nabla \cdot (\mu_A \dot{\gamma}) + H_B \nabla \cdot (\mu_B \dot{\gamma}) + (H_A \rho_A + H_B \rho_B) g + I.T.,$$

$$H_A \frac{D\rho_A}{Dt} + H_B \frac{D\rho_B}{Dt} + (H_A \rho_A + H_B \rho_B) \nabla \cdot u = 0$$

$$H_A + H_B = 1$$



Why Are 3D Free Surface Problems Hard?



Typical problem graph for incompressible flow

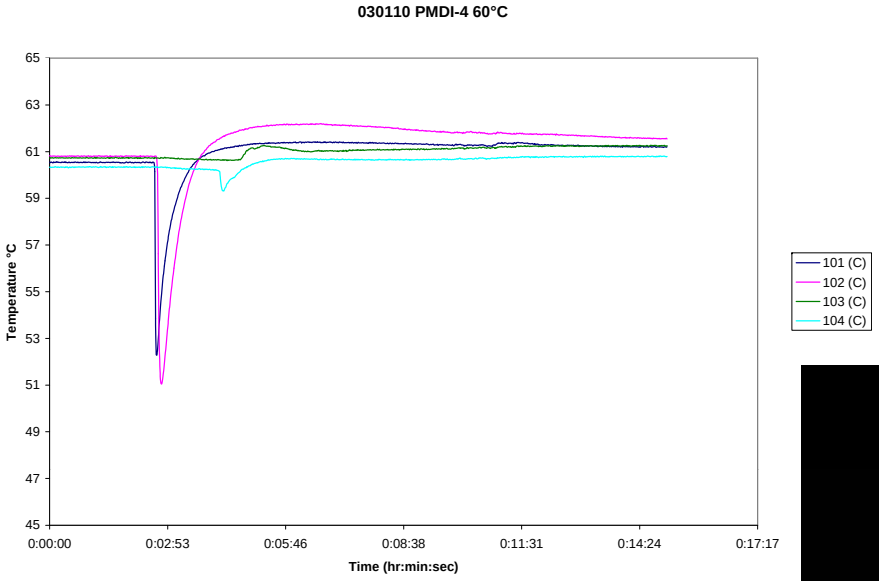
$$\rho \frac{Du}{Dt} = -\nabla P + \mu \nabla^2 v + \rho g$$

$$\nabla \cdot u = 0$$

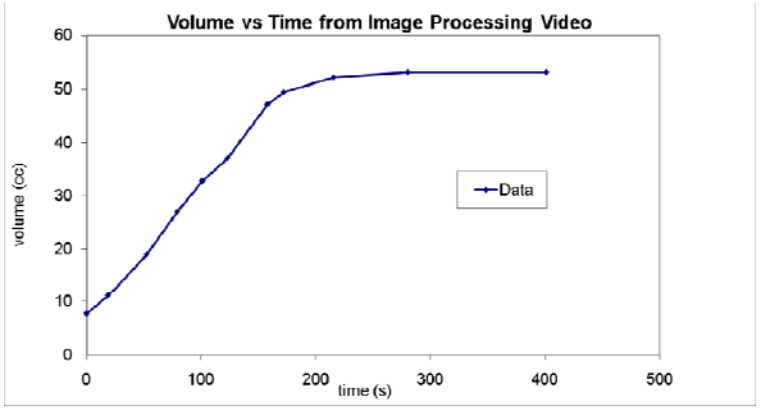
- Formulation uses a coupled u-p solve with a decoupled level set solve
- Incompressibility constraint and distinguishing conditions and boundary conditions lead to non-diagonally dominant matrices
- In 2D, direct solver can be used with LBB elements
- In 3D, only Krylov-based iterative solvers are feasible
- Stabilization for the continuity equation is used to allow for equal order interpolation and improve the matrix condition number
- Stabilized methods that may work well on single phase flows, have difficulty handling the pressure jumps associated with the level set method
- Solution requires heavy duty preconditioner-solver pairing such as ILUT(1-3)/GMRES, which are not very scalable
- Mass loss issues must be ameliorated
    - Remediated via new boundary conditions, stabilization methods, renormalization, and time-stepping algorithm

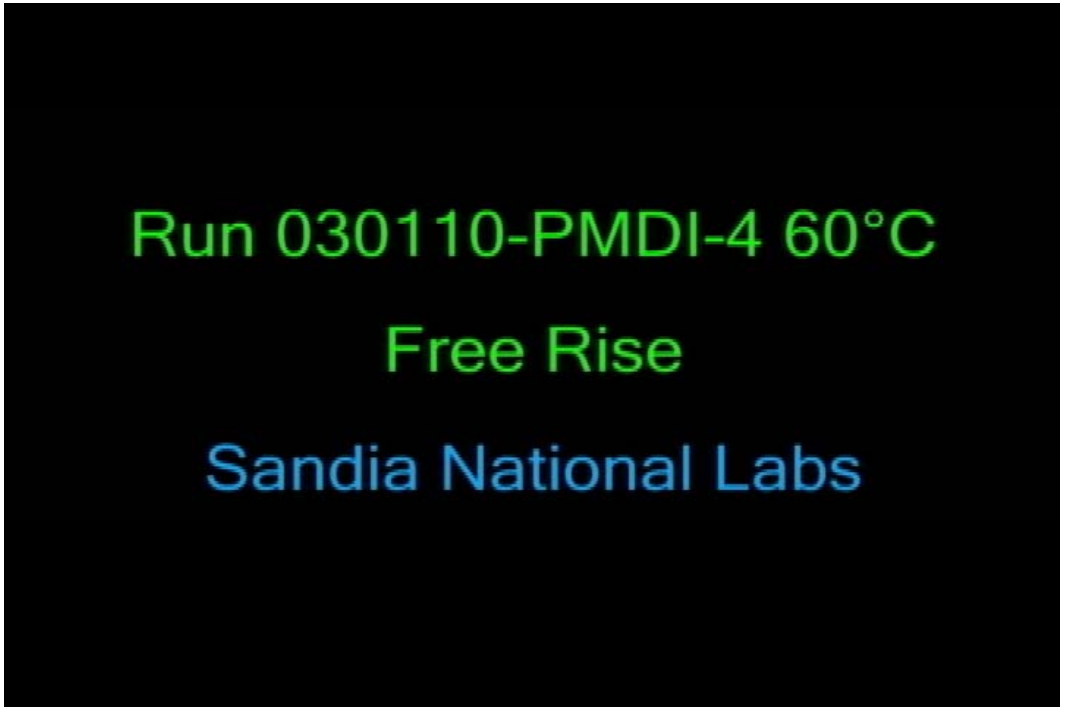
Sandia National Laboratories

PMDI-4 Temperature-Instrumented Flow Viz



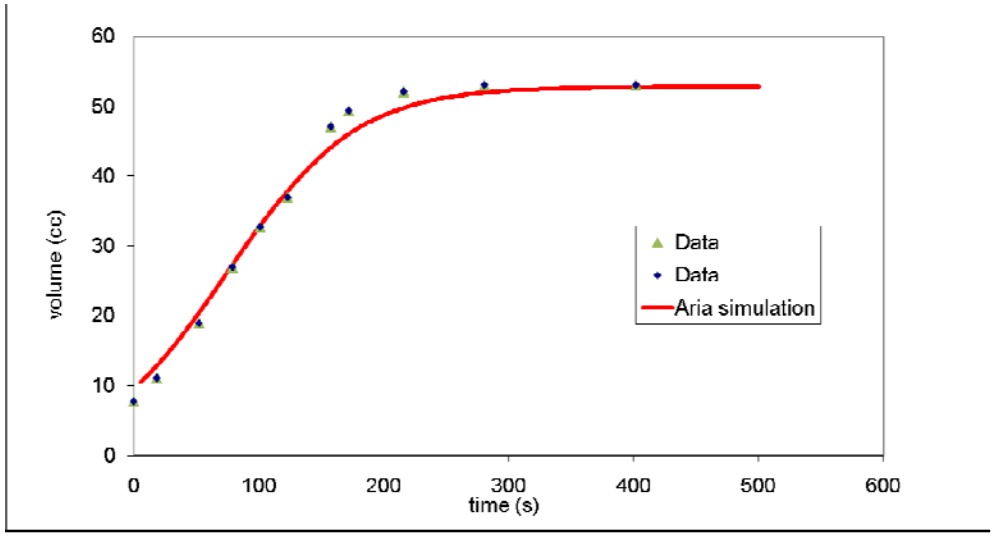
- Front location, Temperatures , Fill rate analysis

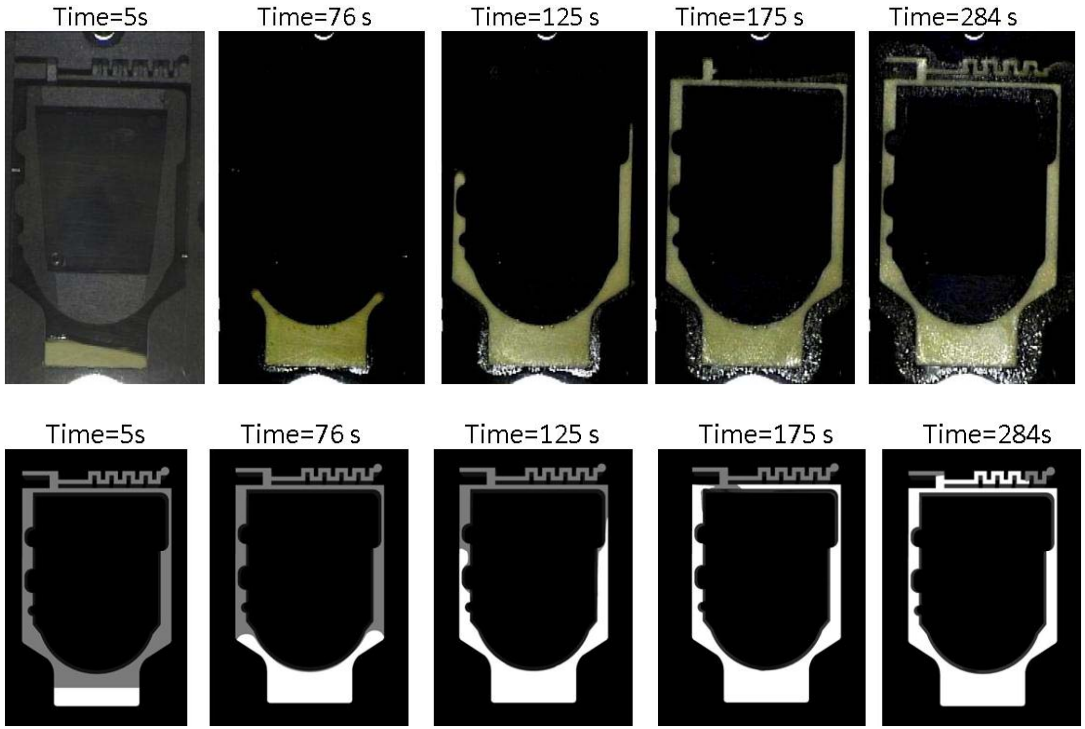




- Video of polyurethane

PMDI-4 Free Surface Validation Study



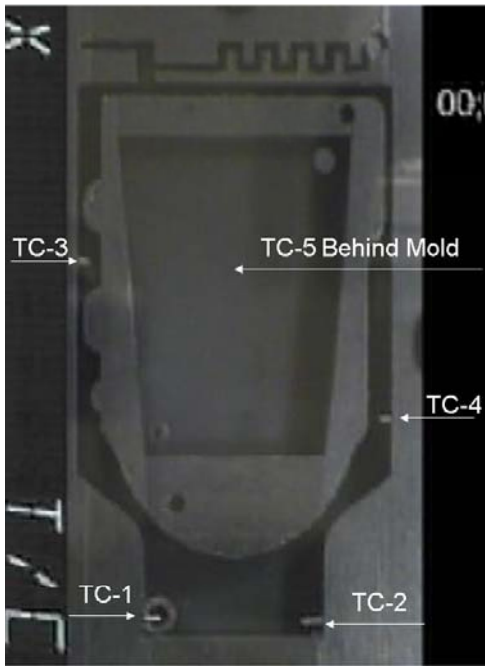


- Model tracks density change from foaming in full system

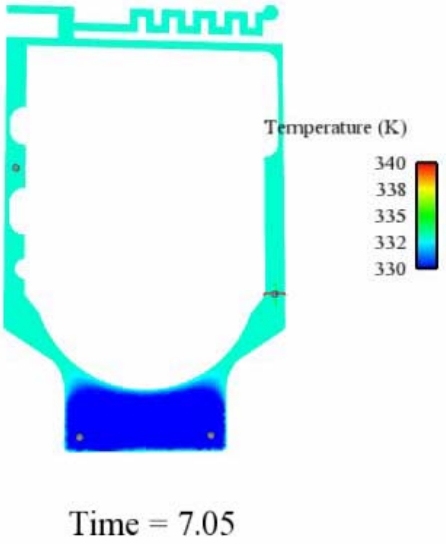
$$\rho = (\rho_i - \rho_f)\exp\left[\frac{-(t-t_i)}{C(T)}\right] + \rho_f$$

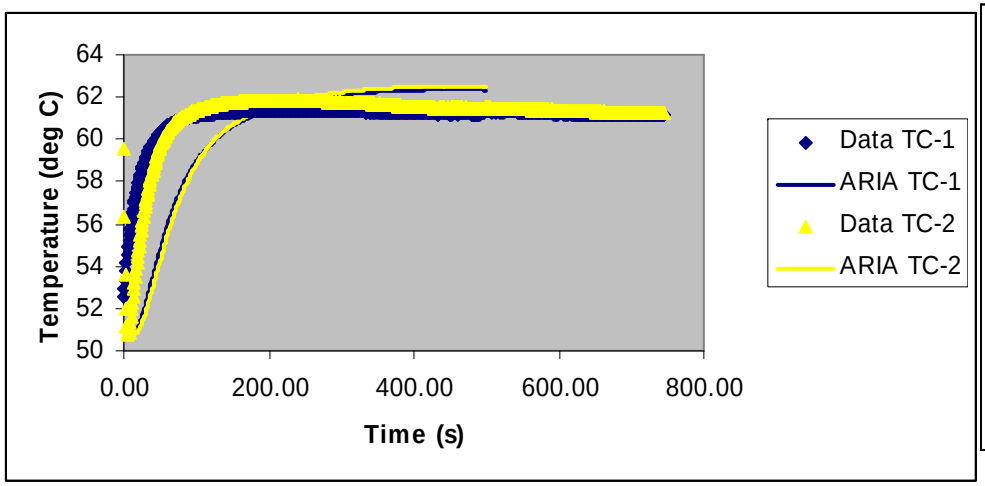
- But validation data show that model foams too slow and then too fast with this simplification
- Current work:
  - Improve density model
  - Add a function to tie foam generation to gelation
  - Work on more complete kinetic model

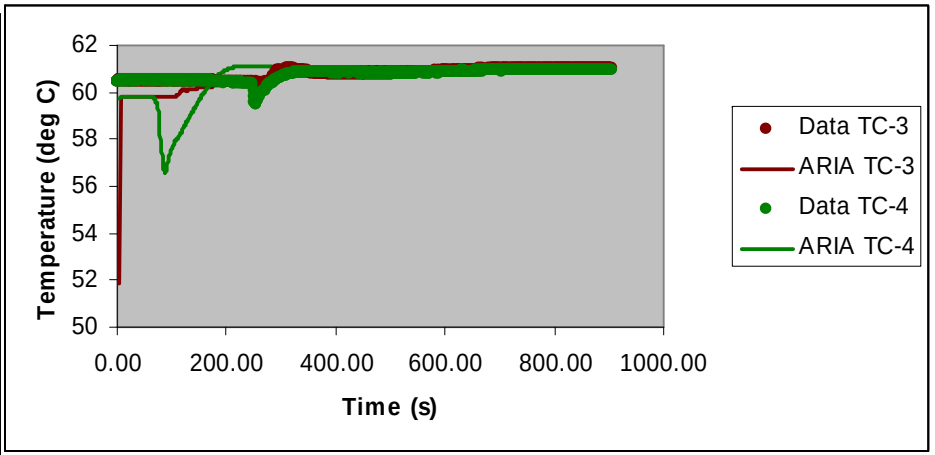
PMDI-4 Thermal Validation Study



- Model captures temperature increase from polymerization exotherm
- Model predicts qualitative trends for various thermocouples

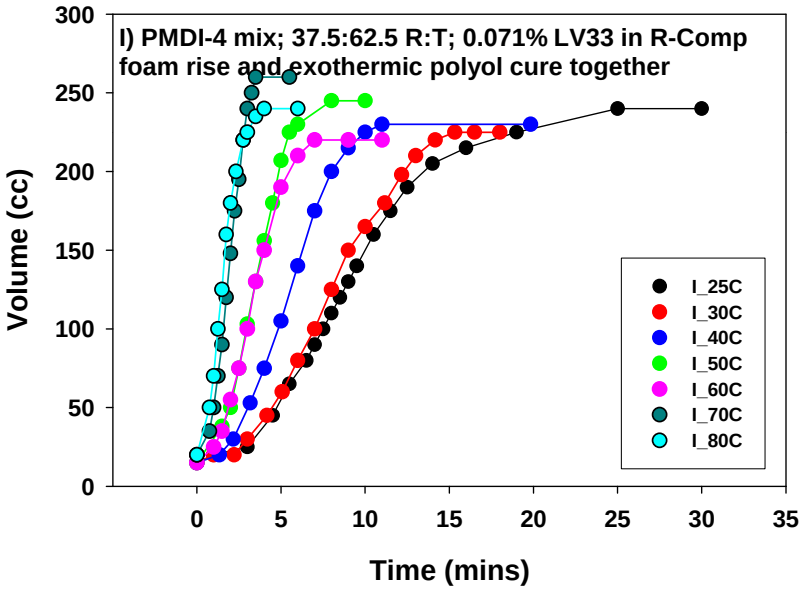


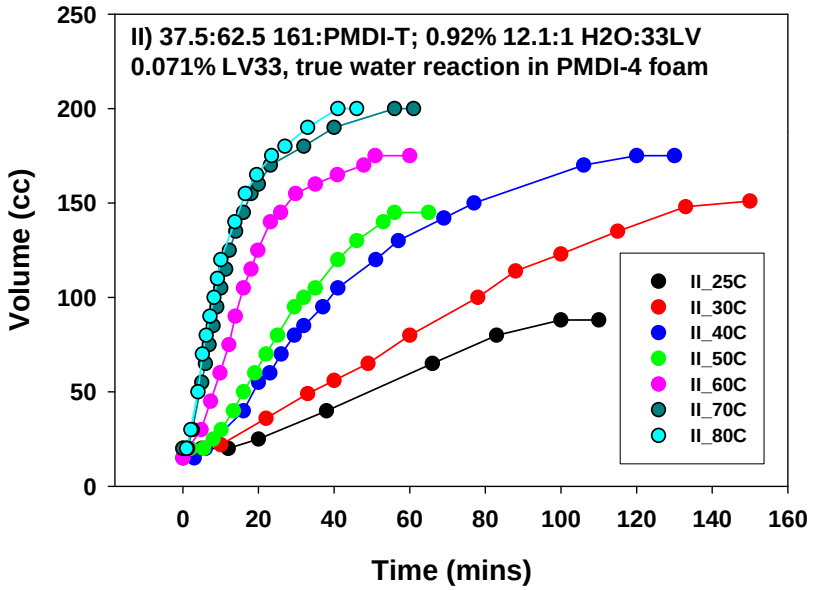




PMDI-4: Improved CO_2 Production Model

- Compare curing PMDI-4 foam with model foam system that does not cure (mid range viscosity epoxy mimics PMDI-4 continuous phase) to deconvolute foaming from increasing viscosity and elasticity, which can lead to bubble pressurization with lack of volume increase

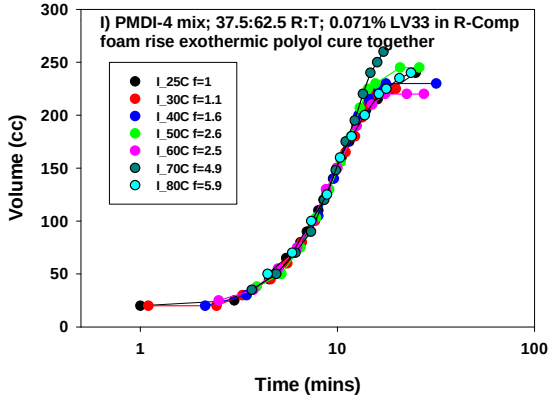


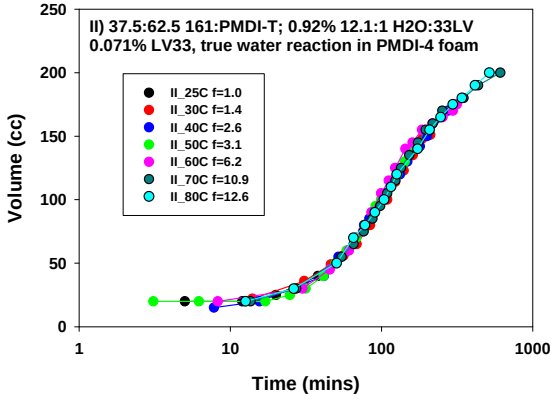


- Curable PMDI-4 foams faster - because of extra heat and synergism from cure reactions? Catalysis of amines?
- The underlying foaming (CO_2 evolution) can be resolved and is much slower

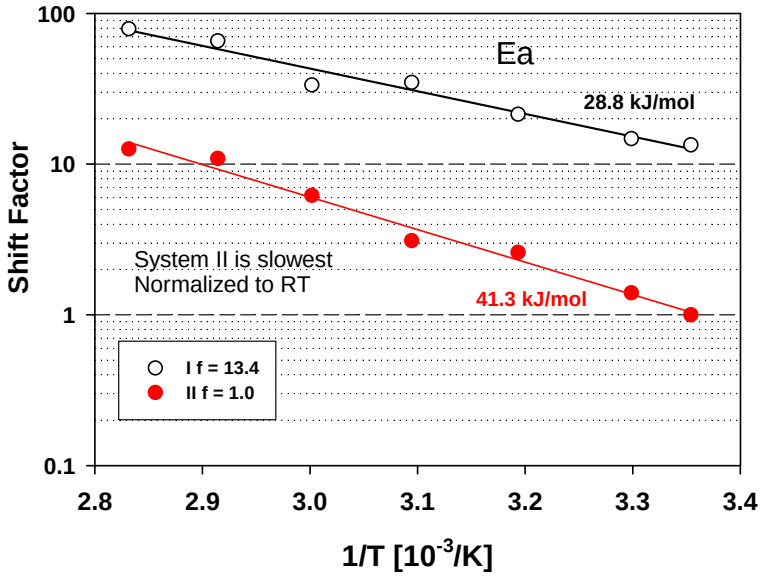
Complex Kinetics of Foaming Reaction

- CO_2 generating foaming reaction due to water-isocyanate has activation energy ΔE ~41kJ/mol
- Curing reactions due to polyol-isocyanate urethane reactions in dried PMDI-4 has roughly the same ΔE ~41kJ/mol
  - The isolated foaming reaction is relatively slow
  - The isolated curing reactions have slightly different rates than in presence of H_20
  - In the presence of polyol (as in the PMDI-4 foam system) we observe much faster foaming action and a different ΔE (29kJ/mol).
  - Not perfectly isothermal due to internal heat of reaction and auto-catalysis?





Superposition of volume-vs-time curves (bottom plot) gives activation energy ΔE with and without curing



Improved Kinetic Model will include CO_2 Generation

$$rate_1 = k_1 e^{-\Delta E_1/RT}[isocyanate]^a[polyol]^b$$ **Polymerization**

$$rate_2 = k_2 e^{-\Delta E_2/RT}[isocyanate]^c[H_2O]^d$$ **CO_2 generation**

- Continue work on kinetics of polymerization coupled to blowing reaction
- Must track five species: water, polyol, polymer, carbon dioxide, and isocyanate , since we have competing primary reaction
- Use existing experiments to determine new Arrhenius rate coefficients

$$\frac{D[CO_2]}{Dt} = +rate_2$$
$$\frac{D[H_2O]}{Dt} = -rate_2$$
$$\frac{D[isocyanate]}{Dt} = -rate_1 - rate_2$$
$$\frac{D[polyol]}{Dt} = -rate_1$$
$$\frac{D[polymer]}{Dt} = +rate_1$$

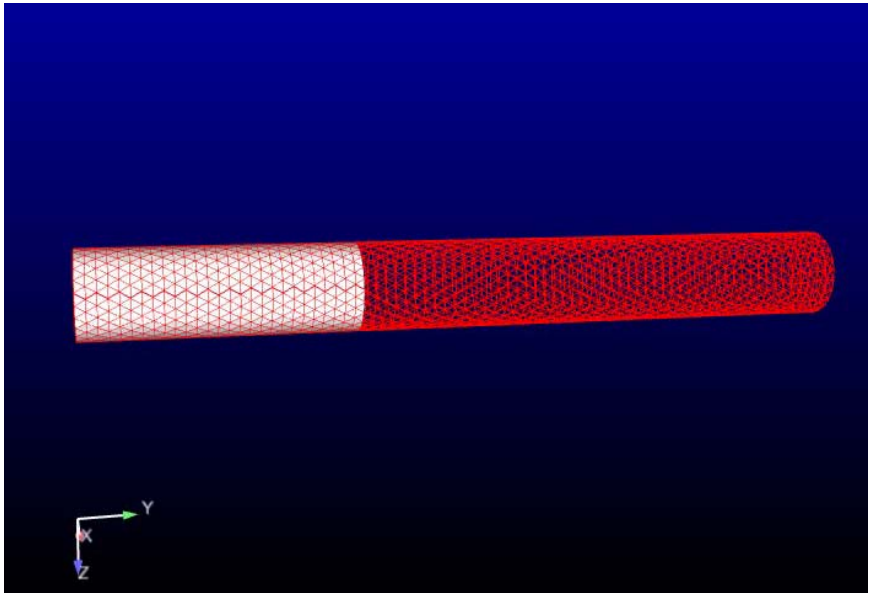
- Must provide initial conditions for all species
- Integrate rate equations as part of the simulation
- Density can now be predicted from gas generation
- **Competing reactions should slow reaction, but actually speeds up foaming while curing is unaffected**

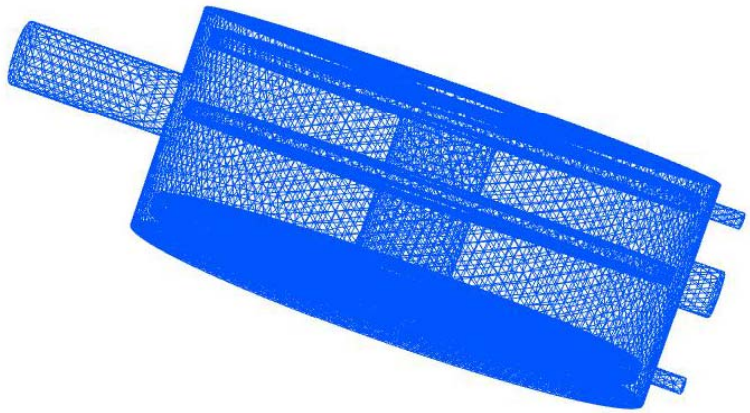
$$\phi(t) = \frac{n_{CO_2}/MW_{CO_2}\rho_{CO_2}}{n_{CO_2}/MW_{CO_2}\rho_{CO_2} + n_{liquid}/MW_{liquid}\rho_{liquid}}$$

$$\rho_{foam} = (\rho_{CO_2} - \rho_{liquid})\phi(t) + \rho_{liquid}$$

Conclusions and Future Work

- Current model is adequate for production calculation *e.g.* determining voids, gate, and vent location
  - Investigate polyurethane encapsulation of new geometries of interest
- Experiments have been completed for advanced model
  - Polymerization and rheokinetics are accurate
  - Need to populate and implement models for more accurate kinetics
- Advanced methods for free surface flows under development
  - Conformal Decomposition Finite Element Method (DR Noble)

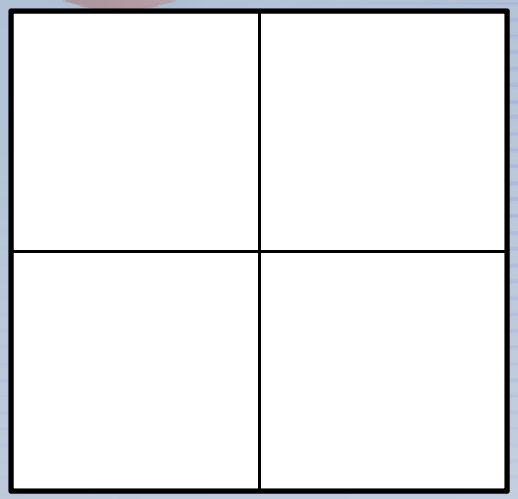




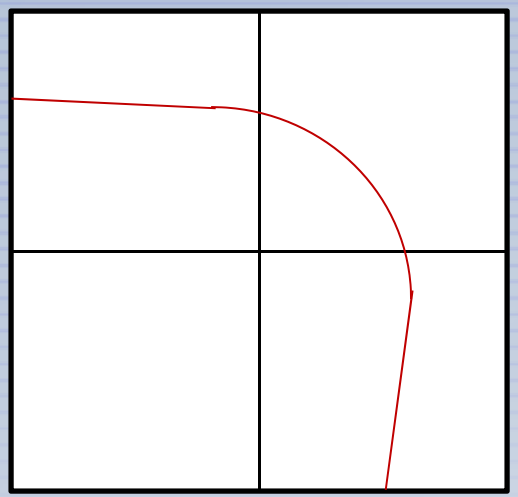
Finite Element Methods for Interfaces in Fluid/Thermal Applications

- **Boundary Fitted Meshes**
  - Supports wide variety of interfacial conditions accurately
  - Requires boundary fitted mesh generation
  - Not feasible for arbitrary topological evolution (ALE)
    - Mesh quality degrades with evolution, phase breakup and merging are precluded.
- **eXtended Finite Element Methods (XFEM)**
  - Dolbow et al. (2000), Belytchko et al. (2001)
  - Successfully applied to numerous problems ranging from crack propagation to phase change to multiphase flow
  - Supports weak conditions accurately, mixed and Dirichlet conditions are actively researched (Dolbow et al.)
  - Avoids boundary fitted mesh generation
  - Supports general topological evolution (subject to resolution requirements)
- **Generalized Finite Element Methods (GFEM)**
  - Strouboulis et al. (2000)
  - Combination of standard finite element and partition of unity enrichment
- **Immersed Finite Element Methods**
  - Li et al. (2003), Ilinca and Hetu (2010)
  - Supports selected jumps across material boundaries (discontinuous gradient or value)
- **Conformal Decomposition Finite Element Method (CDFEM)**
  - Enrichment by adding nodes along interfaces

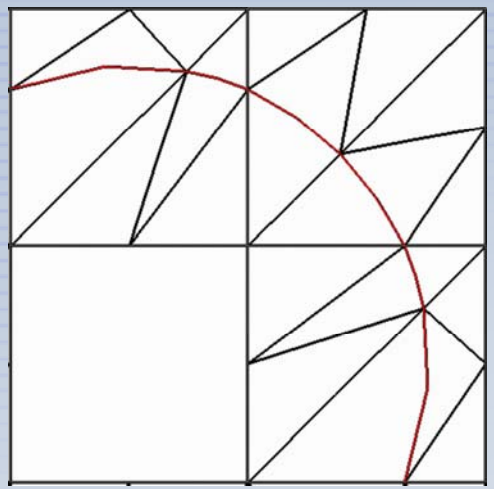
CDFEM Uses Ideas From XFEM, Level Set Methods, and ALE Moving Mesh



Base mesh



Level Set Function



CDFEM Mesh added dynamically at interface

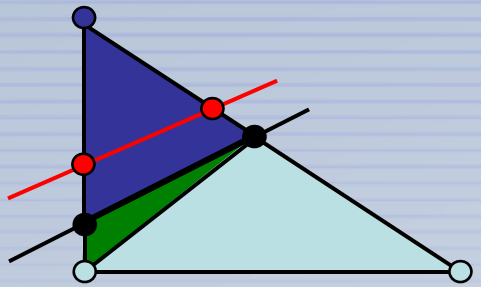
Benefits: Meshed free surface allows for easy application of boundary conditions, discontinuous variables are straight forward, topological changes
Drawbacks: Mass loss similar to diffuse interface methods, expensive, file bloat

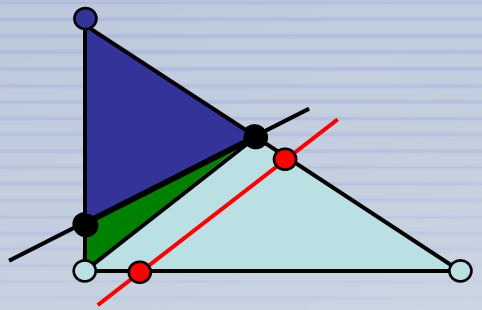
- CDFEM shown convergent for steady flow, Noble et al, IJNMF, 2010
- Extension to moving boundary problems

Sandia National Laboratories

Moving CDFEM

- How do we handle the moving interface?
- What do we do when nodes change sign?
- What space do we use for pressure, velocity and level set?



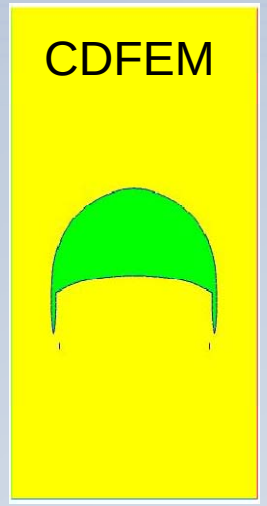


- Goals
  - Try to recover moving mesh case for moving interface
  - Try to preserve minima, maxima
  - Smooth interface
- Proposal
  - Prolongation: Set “old” value to value of nearest point on interface
  - Dynamics: Use ALE style (u-dxdt) for advection term
  - Allow velocity gradient and pressure jumps across interface
  - Level set on sub-element mesh

Code to Code Comparisons for 2D Buoyant Drop: Two Test Problems

Diffuse level set

CDFEM



- Test 2 has fine trailing structures that must be captured by the code
- Density ratio of 1000 and viscosity ratios of 100, Re=35 and Eo=125
- Both CDFEM and a classic diffuse interface method do a reasonable job, but give disparate results
- Results given for coarse mesh (h=1/40)

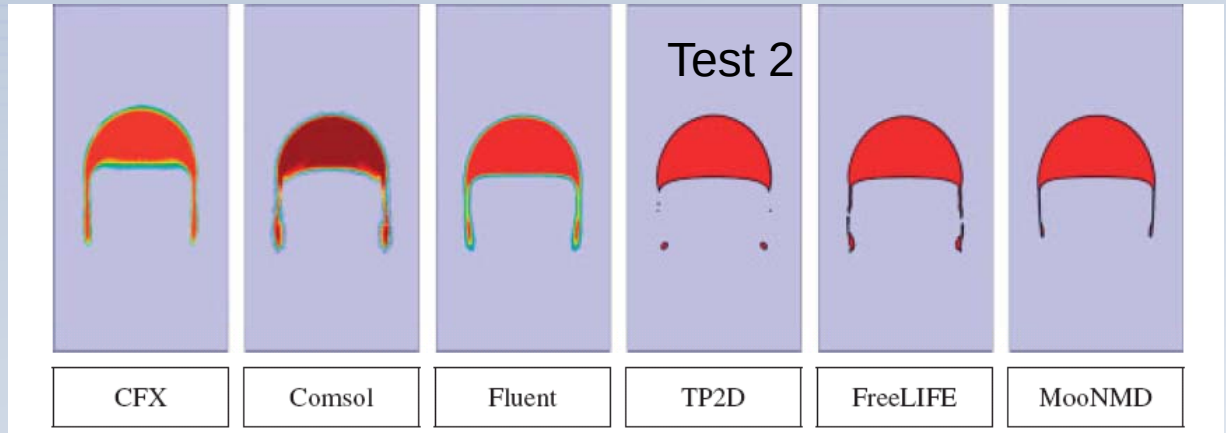
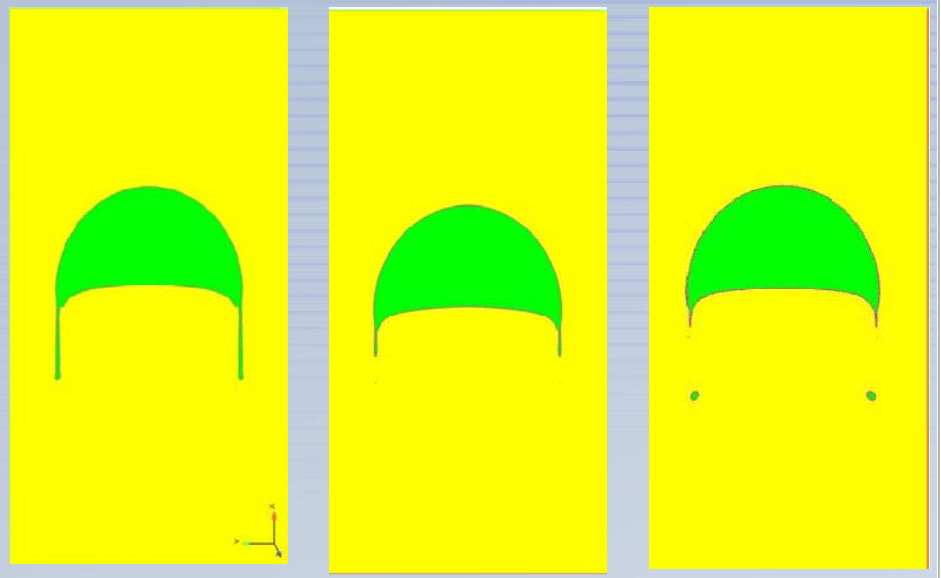
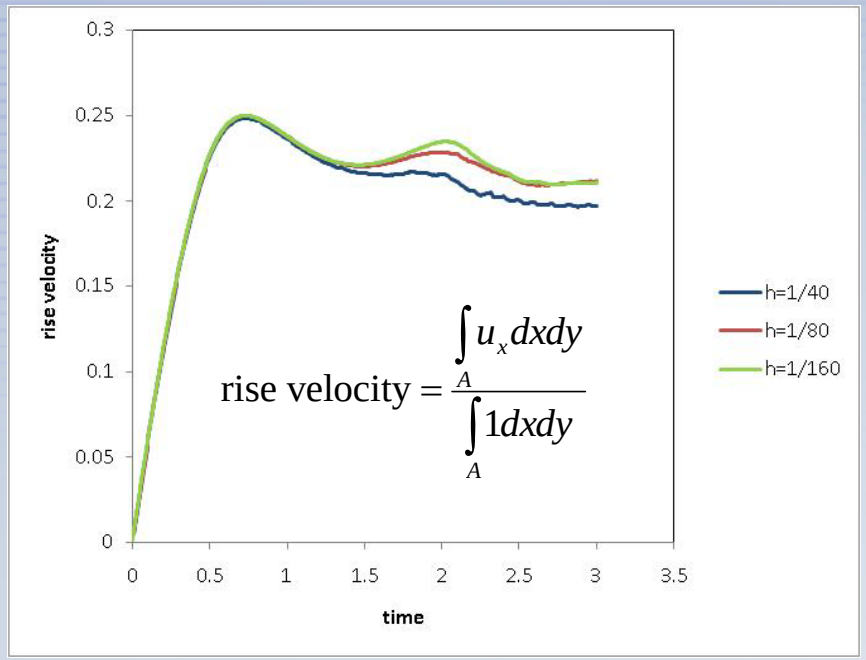


Figure 1. Numerical simulations of a two-dimensional rising bubble for six different codes with identical problem formulations.

Hysing et al, "Quantitative benchmark computations of two-dimensional bubble dynamics, IJNMF, 2009

Mesh refinement study: Constrained CDFEM



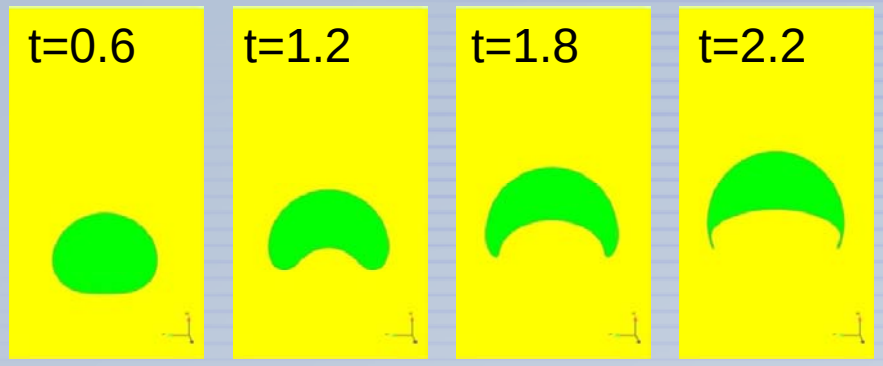


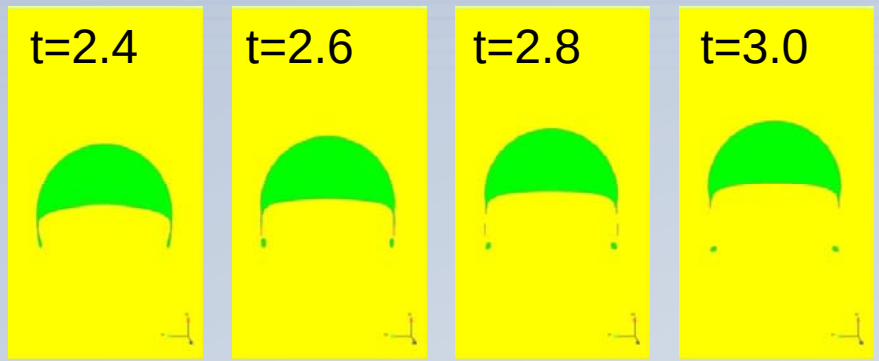
h=1/40 h=1/80 h=1/160

$\Delta t = h/16$

CDFEM with constrained pressure, velocity and level set

Comparison to Hysing et al, 2009





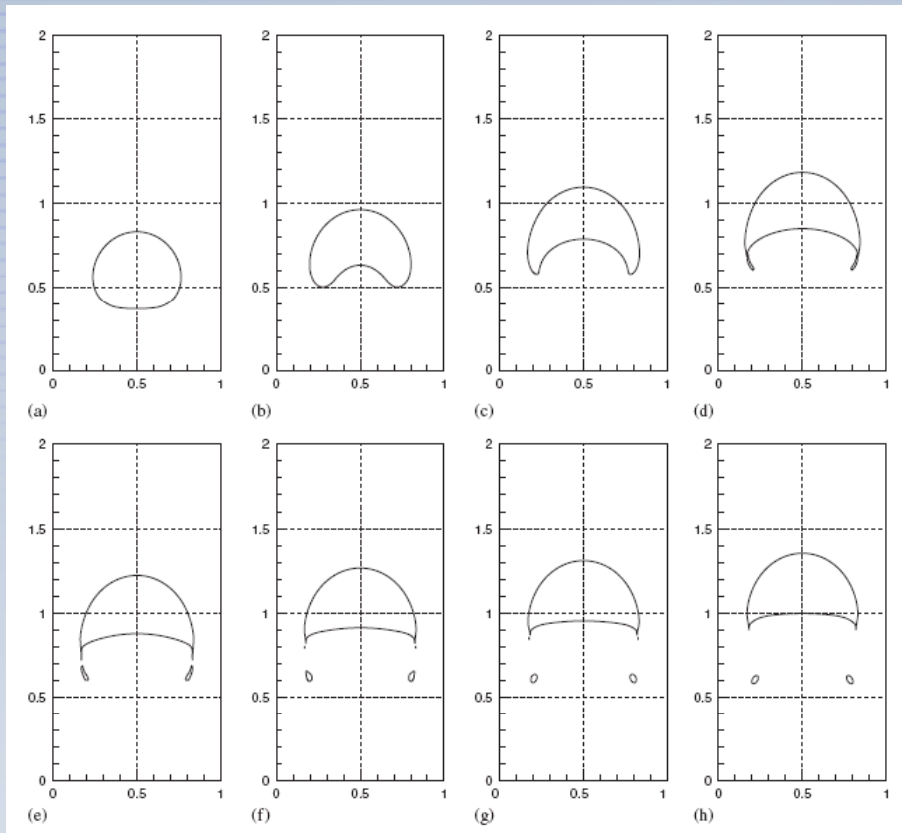
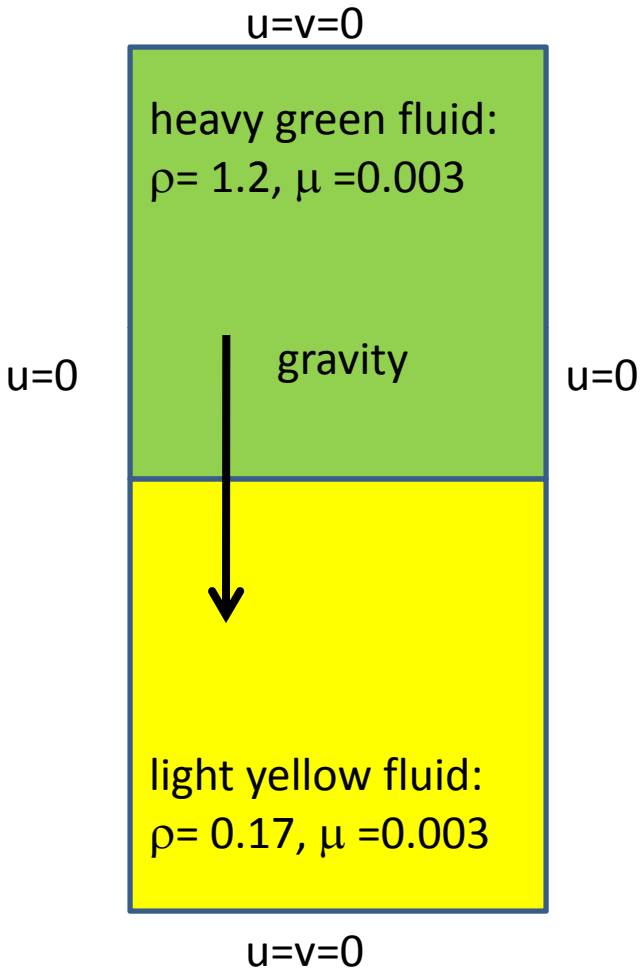


Figure 15. Typical time evolution of the interface for test case 2: (a) $t=0.6$; (b) $t=1.2$; (c) $t=1.8$; (d) $t=2.2$; (e) $t=2.4$; (f) $t=2.6$; (g) $t=2.8$; and (h) $t=3.0$.

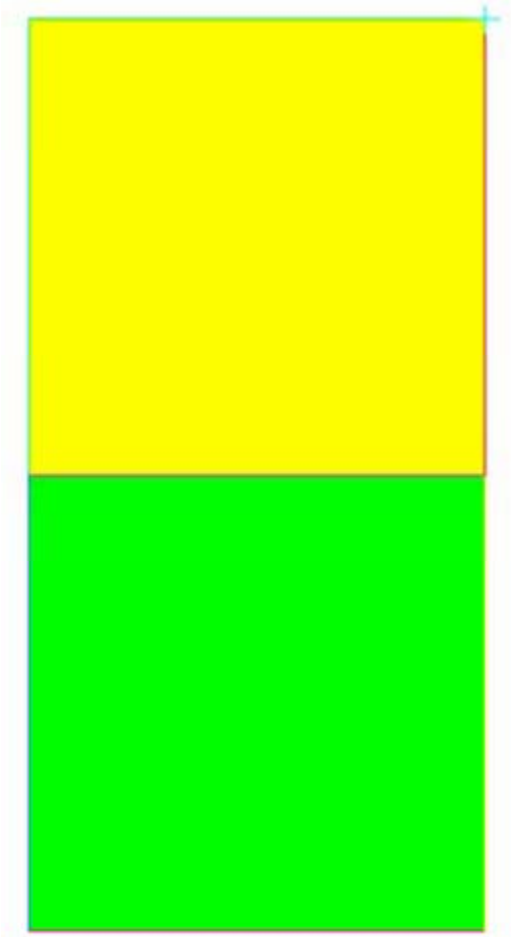
CDFEM with constrained pressure, velocity, and level set, h=1/160

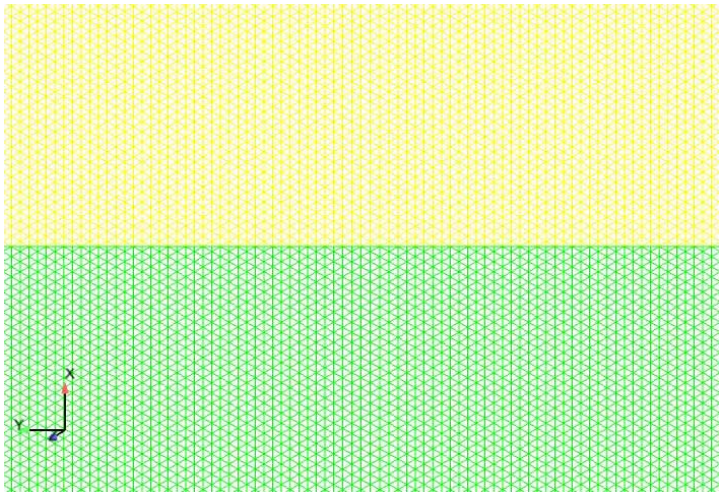
CDFEM: 2D Rayleigh-Taylor Instability



- Unstable stratification of heavy fluid over light fluid initiates Rayleigh-Taylor instability
- Problem similar to Rayleigh-Taylor instability from Smolianski (IJNMF, 2005) but with a 2:1 aspect ratio instead of a 4:1
- Initial condition has flat interface, which will affect wave number of instability
- Results for zero surface tension with fine mesh: h=1/80, dt=h/3

CDFEM: 2D Rayleigh-Taylor Instability

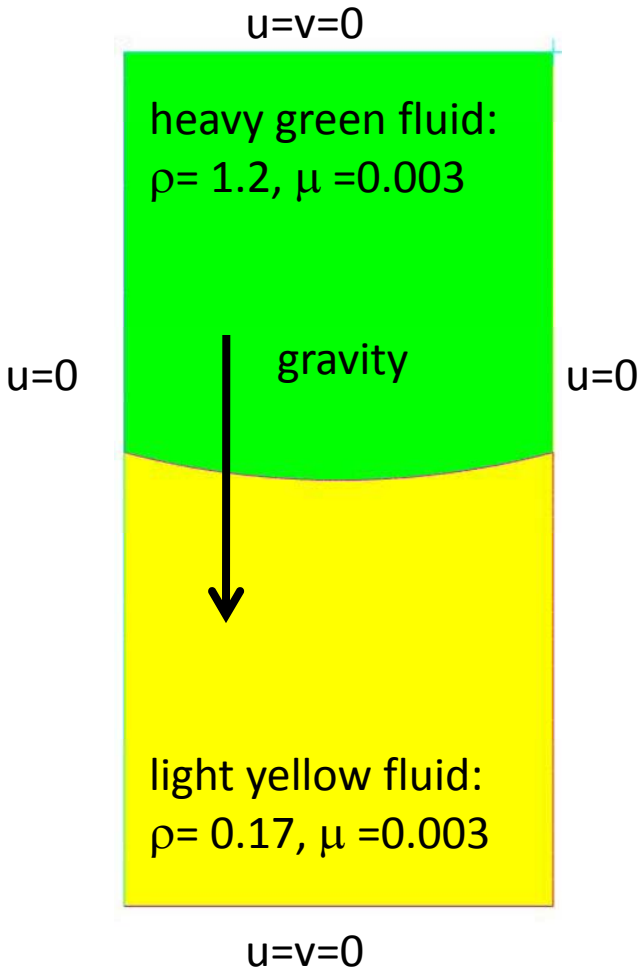




Dynamic CDFEM mesh detail

- Instability initiates from noise in solution and flat interface
- Asymmetric instability form
- CDFEM capture breakup of light fluid as it displaces heavy fluid

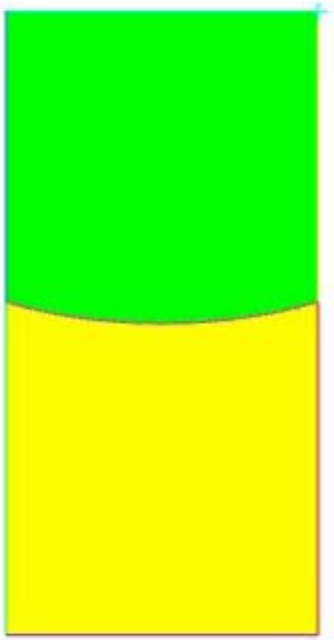
CDFEM: 2D Rayleigh-Taylor Instability



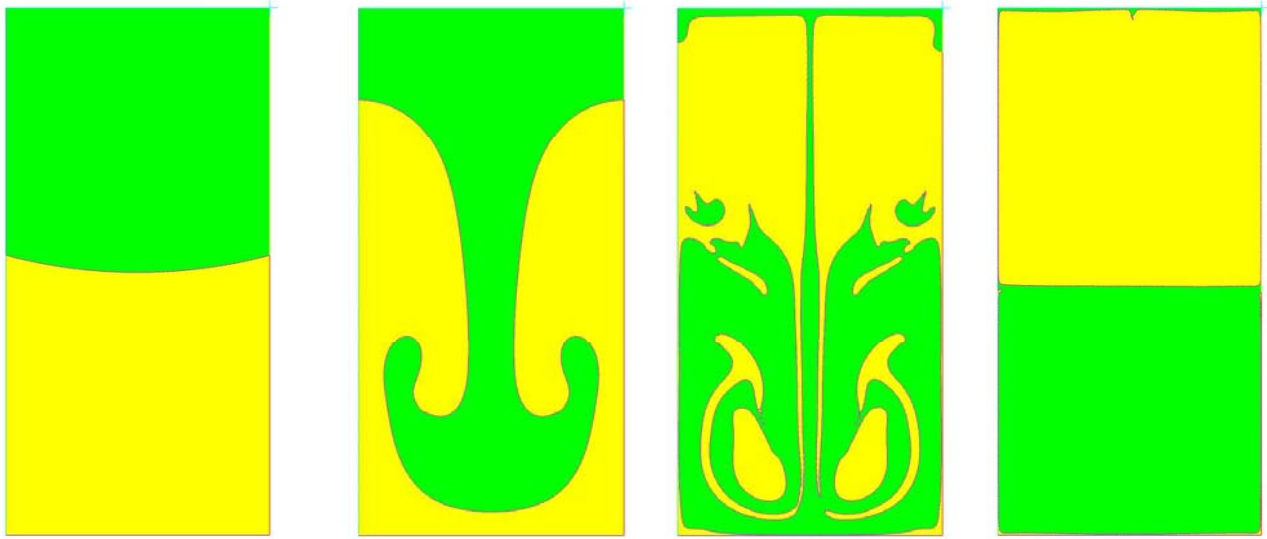
- Unstable stratification of heavy fluid over light fluid initiates Rayleigh-Taylor instability
- Problem similar to Rayleigh-Taylor instability from Smolianski (IJNMF, 2005) but with a 2:1 aspect ratio instead of a 4:1
- Initial condition for the shape of the interface affects wave number and symmetry of instability
- Results for zero surface tension with fine mesh: h=1/80, dt=h/3

- Instability seeded with a perturbation of the free surface
  - Arc of a circle with center = (0,2) and radius 2

Long Time Behavior of Instability



- Symmetric perturbation of the interface
- Symmetric instability
- Long time behavior is a stable state without density inversion
- Mass loss occurs on fine structure, but is less than 10%
- Verification study and documentation in a journal article underway



The End

- Thank you for your attention
- Questions, comments, etc?