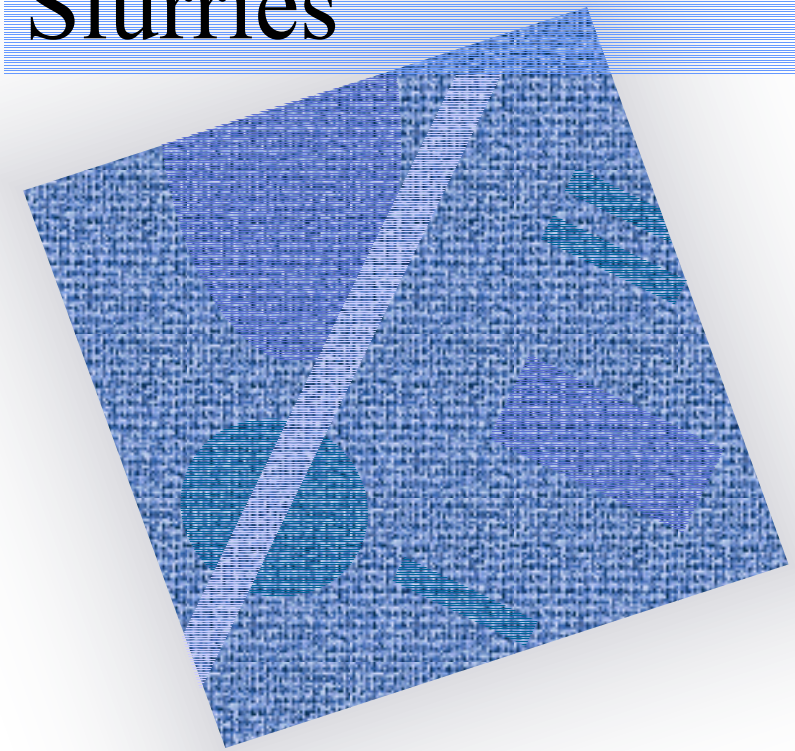
Mechanics of Bubbles in Sludges and Slurries



PA Gauglitz, G Terrones, and DP Mendoza

PNNL

MM Denn, SJ Muller, and BT Liu

LBNL, UC Berkeley

WR Rossen SI Kam

UT Austin

Environmental Management Science Program - DOE

Introduction

The Hanford Site in Washington has 177 underground storage tanks containing radioactive wastes that are complex mixes of radioactive and chemical products. Previous studies have established that the waste surface level in a tank responds to barometric pressure changes, the compressibility of retained bubbles accounts for the level changes, and the volume of retained gas can be determined from the measured waste level and barometric pressure changes. However, interactions between the gas bubbles and rheologically complex waste are not well understood and result in inaccurate estimates of retained gas.

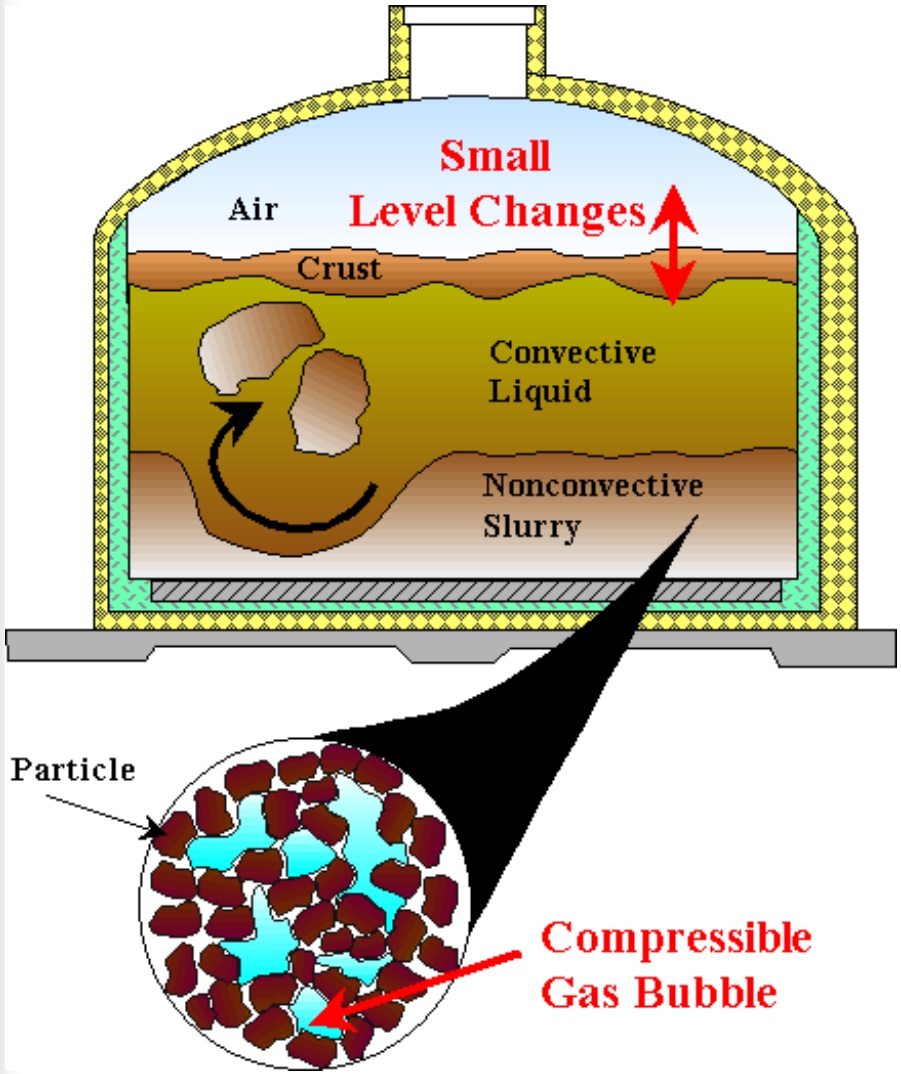
Because the retained gas is typically a flammable mixture of hydrogen, ammonia, and nitrous oxide, accurate determination of its volume is critical for establishing the safety hazard of the tanks. Accurate estimates of retained gas from level/pressure data are highly desirable because direct in situ measurements are very expensive in an individual tank and impossible in many tanks.

AW Tank Farm

Motivation

- Flammable Gas Hazard Is Widespread at Hanford
  - Gas generation always occurs (widely varying rates)
  - Retention is known to occur in many, but not all tanks
- Flammable Gas Hazard Impacts All Waste Operations
  - Safe storage
  - Salt-well pumping
  - Retrieval and pretreatment
- Retained Gas Volume Can Be Estimated from Waste Compressibility
  - Surface level/barometric pressure correlation

Flammable Gas Hazard Depends on Amount of Retained Gas



- Sometimes a large volume of flammable gas is retained; a key issue is how much gas.
- The gas can burn if a large and rapid release occurs together with an ignition source.
- Barometric pressure changes cause small expansion or compression of bubbles and level changes. Amount of retained gas can be estimated from level changes.

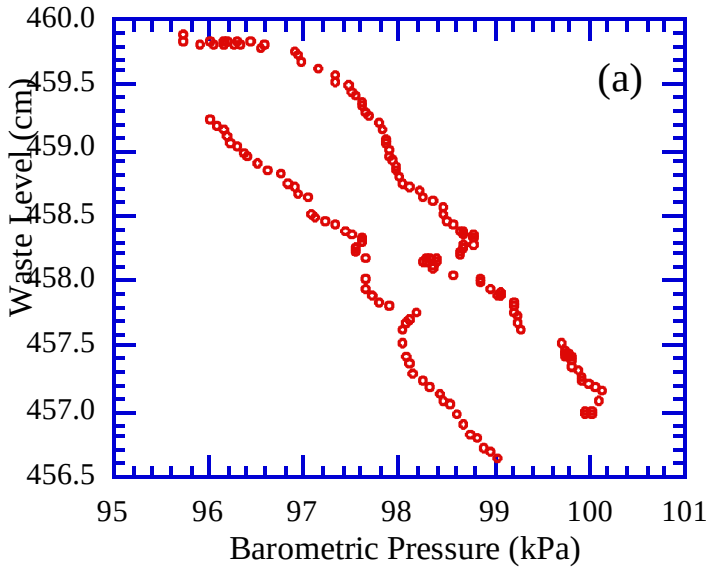
Overall Objectives

- Understanding behavior of single bubbles in “soft solids”
- Develop a framework for predicting macroscopic behavior from the underlying single-bubble behavior

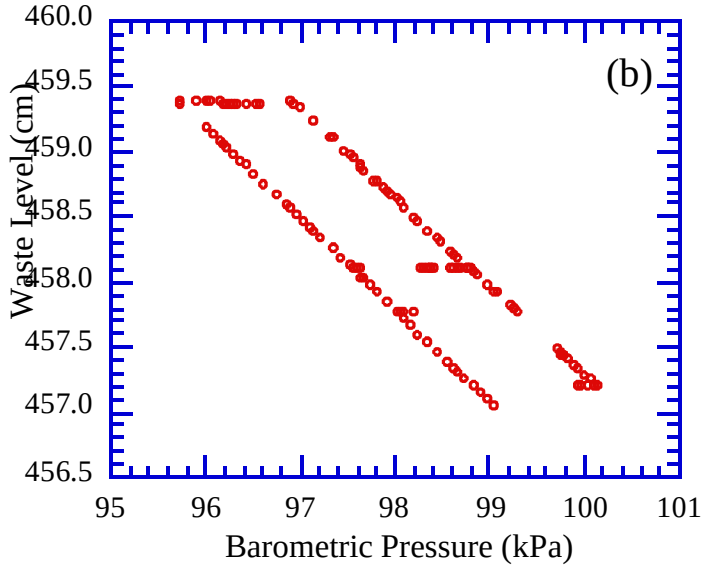
Applications

- Improving the accuracy of gas volume estimates
- Provide more accurate models for estimating waste properties from level/pressure data
- Quantify the effect of barometric pressure fluctuations on the slow release of bubbles

Waste Level Versus Barometric Pressure for Tank S-106



Measurements



Elastic-Plastic Model

From Whitney et al. (1996)

Understanding the primary mechanisms of gas bubble retention in wastes is critical because they define the dominant forces acting to retain the bubbles, and these forces will be among the dominant forces affecting the bubbles during small expansions and contractions caused by barometric pressure fluctuations. A series of studies has focused on the mechanisms of bubble retention (Gauglitz et al. 1994, 1995, 1996; Rassat et al. 1997), and the predominant retention mechanisms are bubbles retained by the strength of the waste material and bubbles retained by capillary forces.

The adjacent figure shows images of bubbles retained by waste strength in sludge-like material from Hanford Tank S-102 (Gauglitz et al. 1996). For comparison, one image shows the shape of bubbles retained in a bentonite clay simulant with properties that mimic some properties of the actual waste material. In many ways, the bubbles in the bentonite clay are similar to the retained bubbles in the actual waste material. Still, bentonite clays do not mimic all of the behavior necessary for studying bubble expansion and compression in sludge simulants.

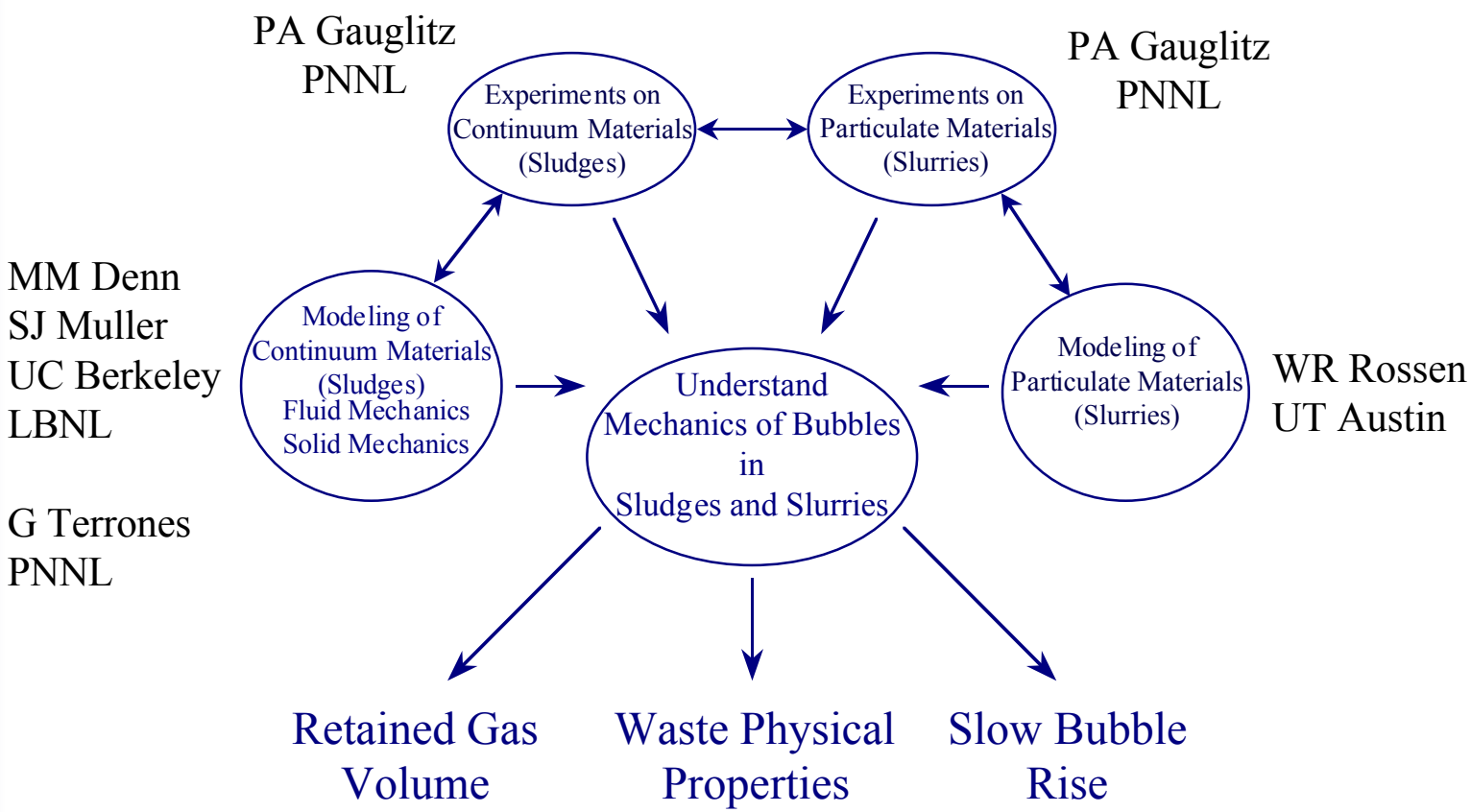
Images of Retained Bubbles

241-S-102
"Stiffer Sludge-Like"
Material

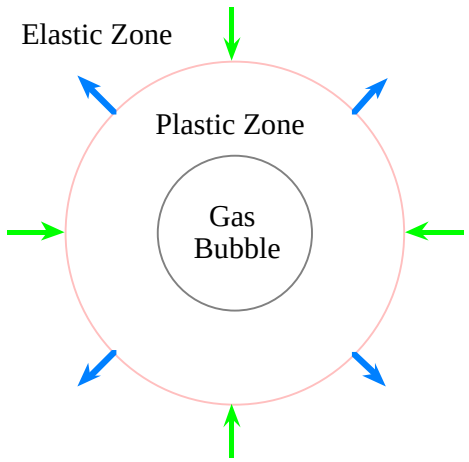
241-S-102
"Softer Sludge-Like"
Material

Bentonite Clay
Simulant

Collaborative EMSP Project PNNL, LBNL, and UT Austin



Solid Mechanics Modeling



Stress and Strain Fields

Elastic Zone

Equilibrium and Compatibility Equations

Plastic Zone

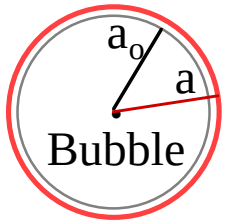
Equilibrium and Yield Criterion (Expansion and Compression) Equations

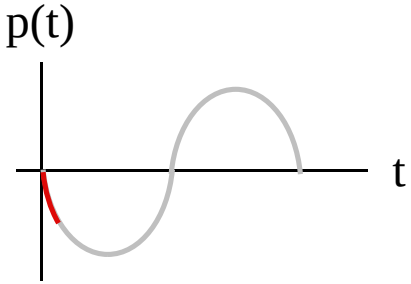
- Bubble Imbedded in Elasto-Plastic Medium
- Quasi-Static Periodic Variation of External Pressure
- Von Mises Yield Criterion
- Solutions are Matched at the Plastic-Elastic Boundaries
- Residual Stresses are Taken into Account
- Logarithmic Strains used to Calculate Finite Plastic Displacements

Beginning of External Decompression

$$P(t) = p_o + p(t)$$

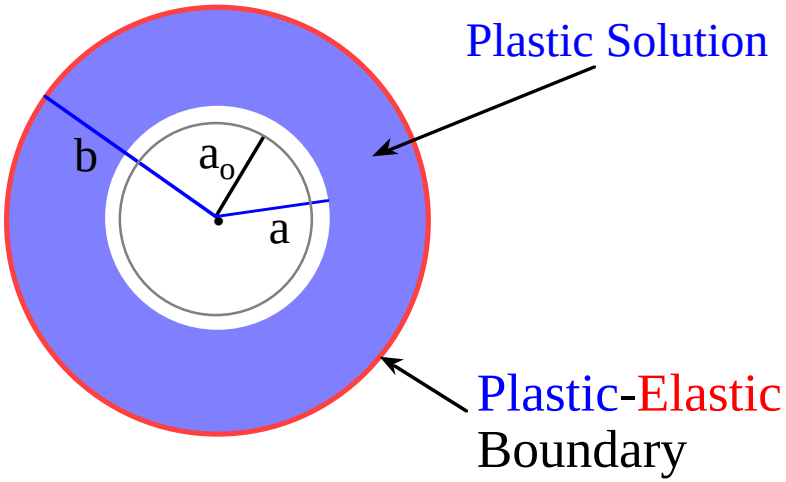
Elastic Solution Everywhere

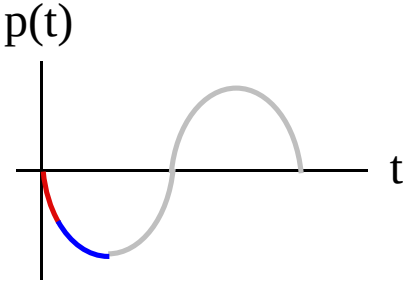




The Yield Criterion is Satisfied within a Finite Region of the Solid Medium

Elastic Solution

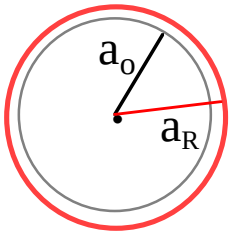


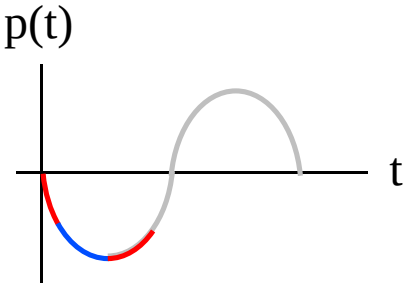


Beginning of Compression Sweep

Initial Condition is a State of Residual Stresses

Elastic Solution Everywhere

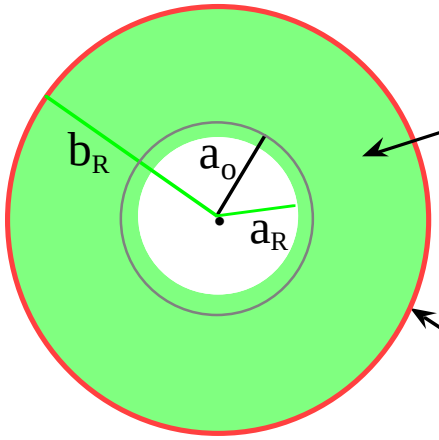




The Reversed Yield Criterion is Satisfied within a Finite Region

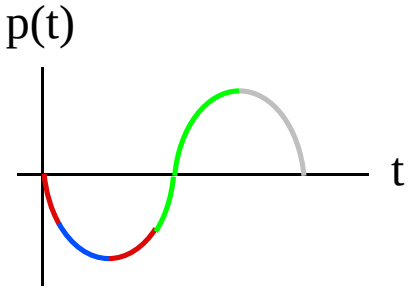
Elastic Solution

Reversed Plastic Zone Solution

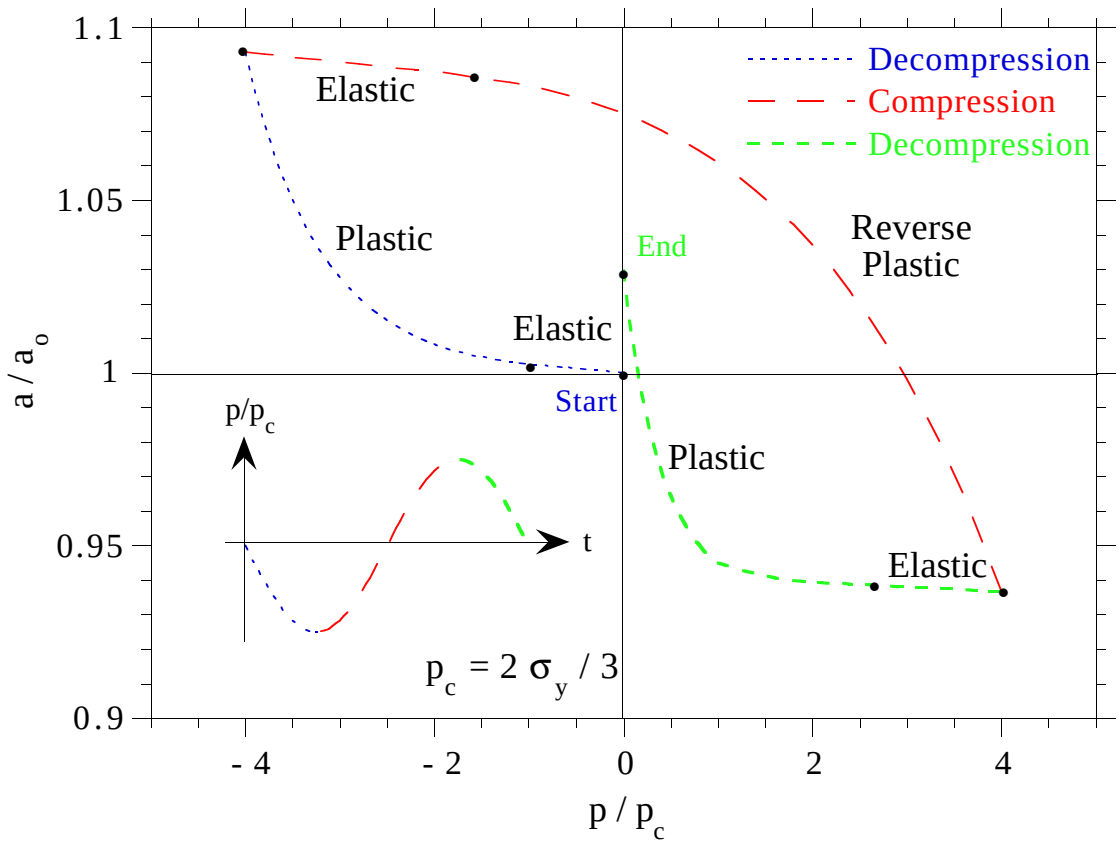


Reversed Plastic-Elastic Boundary

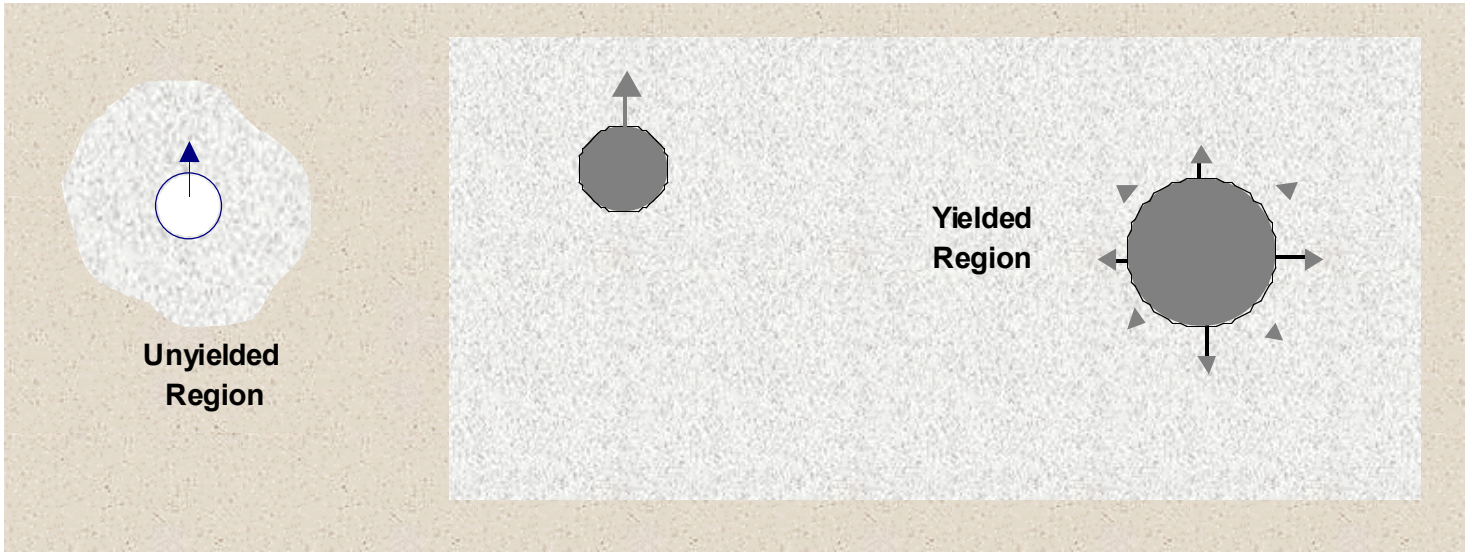
$b_R < b$



Modeling Results Show Hysteresis



Continuum Modeling of Sludges

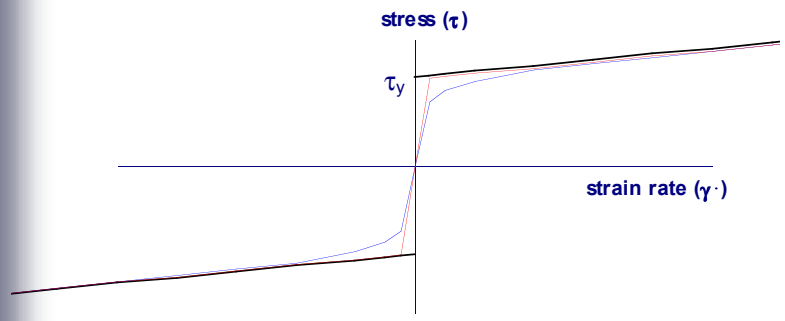


- The sludge surrounding the bubbles will be treated as a single continuous medium (a yield stress fluid) that includes both yielded and unyielded regions
  - How do barometric pressure changes and bubble expansion affect bubble motion and the yielded region?
  - How do yielded regions surrounding adjacent bubbles interact?

Constitutive Relationships for Yield Stress Fluids

Continuum modeling requires a relationship (a constitutive equation) that describes the state of stress at any point

1D Constitutive Relationships



Bingham Material

$$\underline{\underline{\tau}} = \overline{\eta}_0 + \frac{\tau_y}{\|\dot{\gamma}\|}\underline{\underline{\dot{\gamma}}} \qquad \|\tau\| \ ? \ \tau_y$$

$$\underline{\underline{\tau}} = G\underline{\underline{\gamma}} \qquad \|\tau\| < \tau_y$$

Regularization

$$\underline{\underline{\tau}} = \overline{\eta}_0 + \frac{\tau_y}{(\|\dot{\gamma}\| + \varepsilon)}\underline{\underline{\dot{\gamma}}}$$

Biviscosity Model

$$\underline{\underline{\tau}} = \overline{\eta}_0 + \frac{\tau_y}{\|\dot{\gamma}\|}\underline{\underline{\dot{\gamma}}} \qquad \|\dot{\gamma}\| \ ? \ \dot{\gamma}_c$$

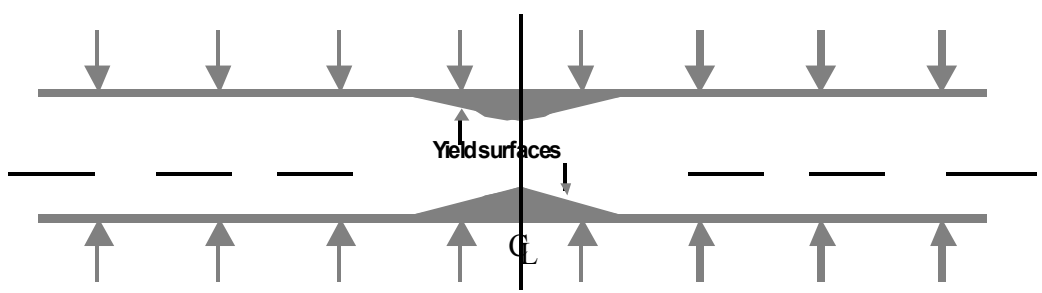
$$\underline{\underline{\tau}} = \eta_1\underline{\underline{\dot{\gamma}}} \qquad \|\dot{\gamma}\| < \dot{\gamma}_c$$

- The simplest constitutive equation for a yield stress fluid is that of a Bingham material
  - Discontinuity at zero strain rate presents problems for numerical implementation
  - Elastic response prior to yield ($\|\tau\| < \tau_y$) usually neglected ($G \rightarrow \infty$)
- The Bingham material has been approximated using continuous equations, including a regularization parameter model and a biviscosity model

Yield Surfaces

- Determining yield surface locations is critical in modeling yield stress fluids
  - A yield surface separates yielded and unyielded regions
    - Yielded regions act as viscous fluids
    - Unyielded regions act as elastic solids
  - Whether the material is yielded at any given point is determined by the state of stress at that point

- Axisymmetric squeeze flow, an elementary prototype for bubble expansion and contraction, is shown below (unyielded region indicated in gray)
  - Motion and interactions of yield surfaces in complex geometries are not understood

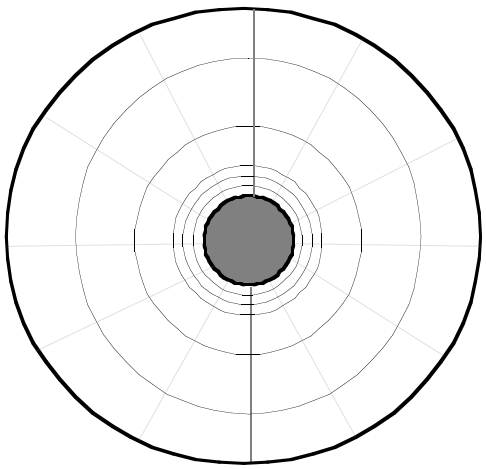


Numerical Implementation

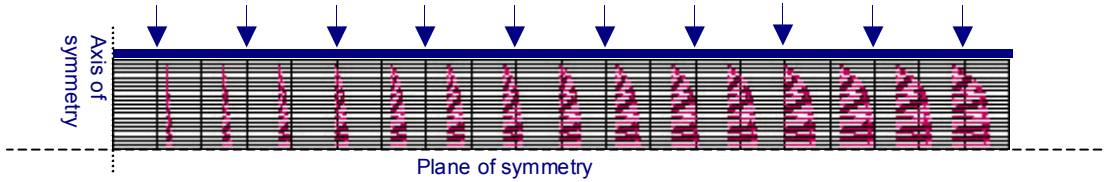
The constitutive equation, along with the equations of momentum and mass conservation, yields a set of governing nonlinear partial differential equations that cannot be solved analytically.

- The finite element method is used
  - A regularized Bingham equation is used
  - The domain is divided into a mesh of discrete subdomains
  - An approximate solution is sought that satisfies the governing equations in a weighted integral sense
  - The resulting system of equations is solved for an approximate solution over the entire domain

Sample Mesh for Bubble Simulation

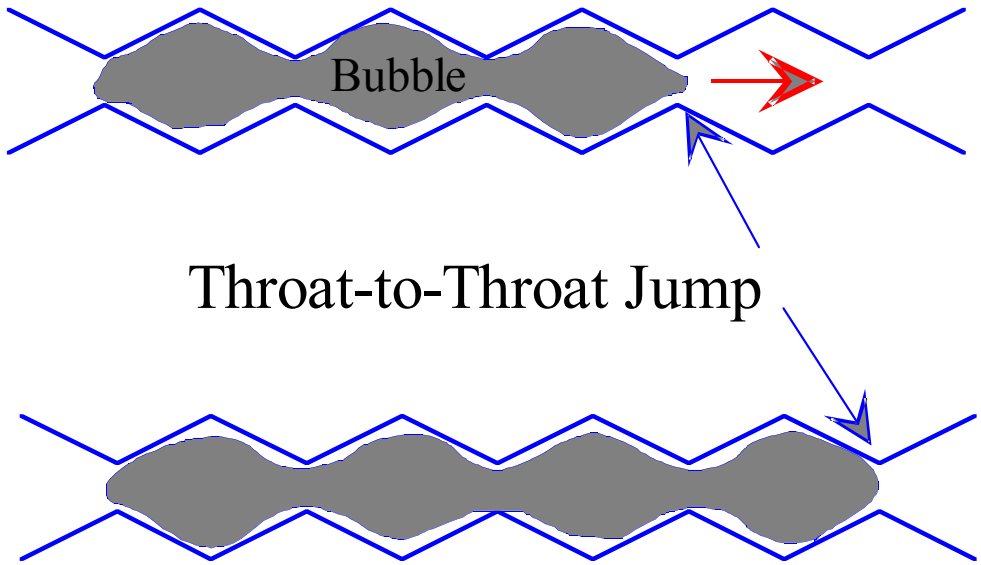


Mesh for axisymmetric squeeze flow (calculated velocity field shown in red)



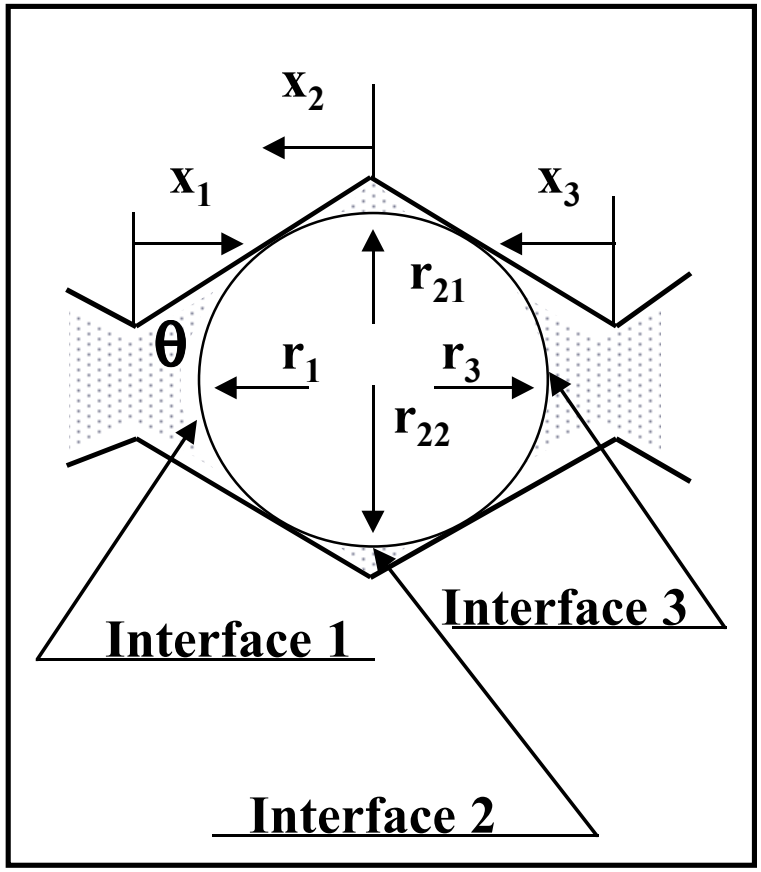
Bubble Behavior in Particulate Materials

Pore-Level Capillary Hysteresis



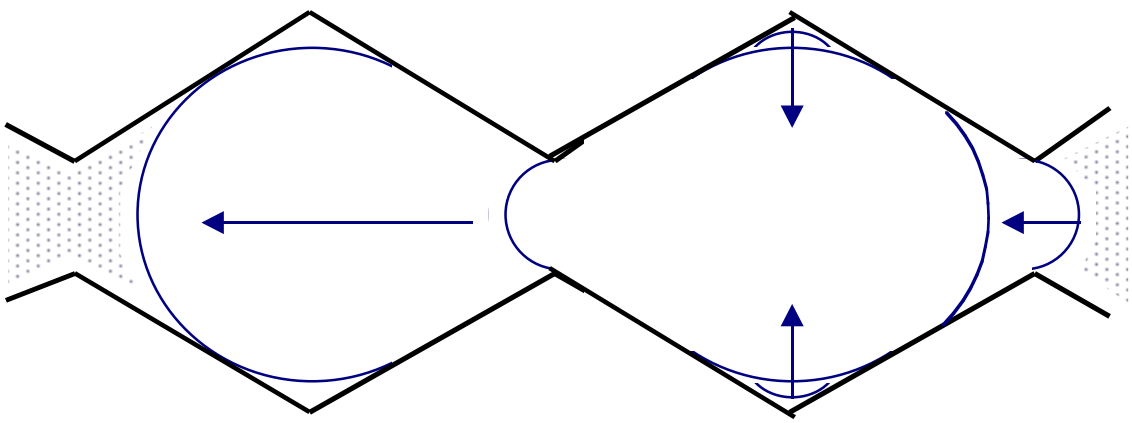
Modeling Bubble Growth in Particulate Materials

- Three Interfaces:
  - Two at pore throats (interfaces 1, 3)
  - One at pore body (interface 2)
- Interface position x_i determines radius of the corresponding interface i for given contact angle θ .



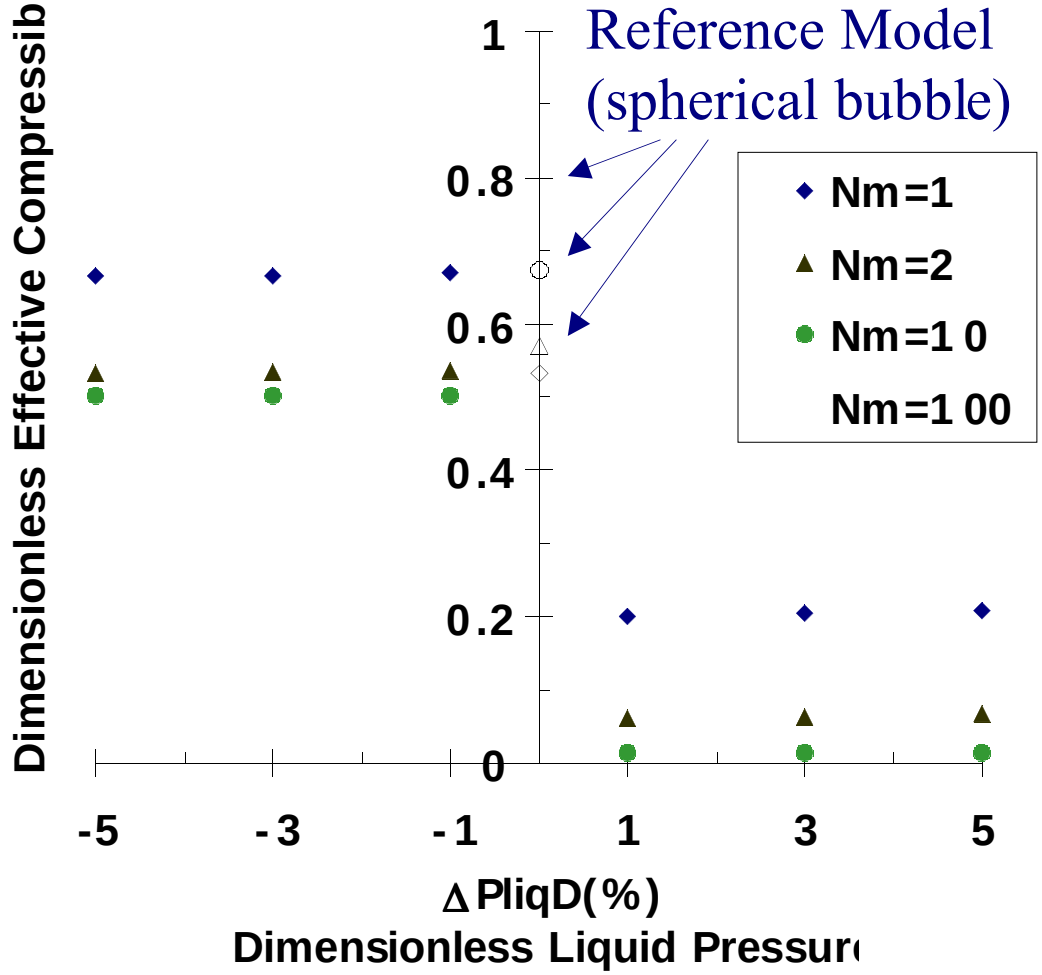
Modeling Bubble Growth

- Bubble grows until it reaches maximum capillary pressure ($P_{c,\,max}$ or P_c^e) as bubble front moves into the pore throat.
- Position (or angle) of advancing interface determines the other variables due to capillary equilibrium.
- Because of instability at $P_{c,\,max}$, only one interface passes through pore throat; other interface retreats.

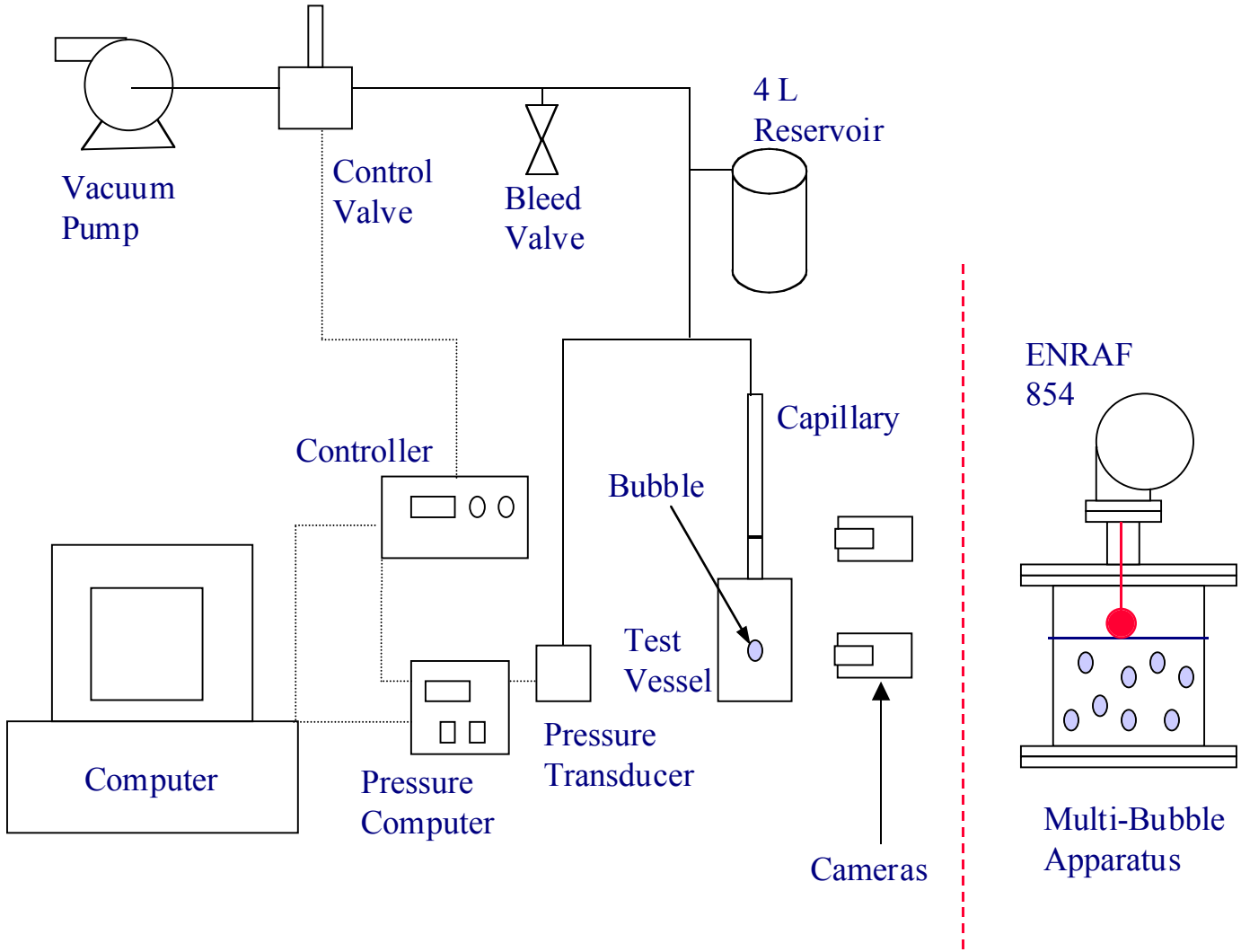


Bubble Compressibility Depends on Direction of Pressure Change

Base case : $\theta = 0^o$, $P_{liq} = P_c^e$, $R_b/L = 0.5$, $R_b/R_t = 5$



Apparatus for Single Bubble Studies



Transparent Simulants for Studying Bubble Behavior

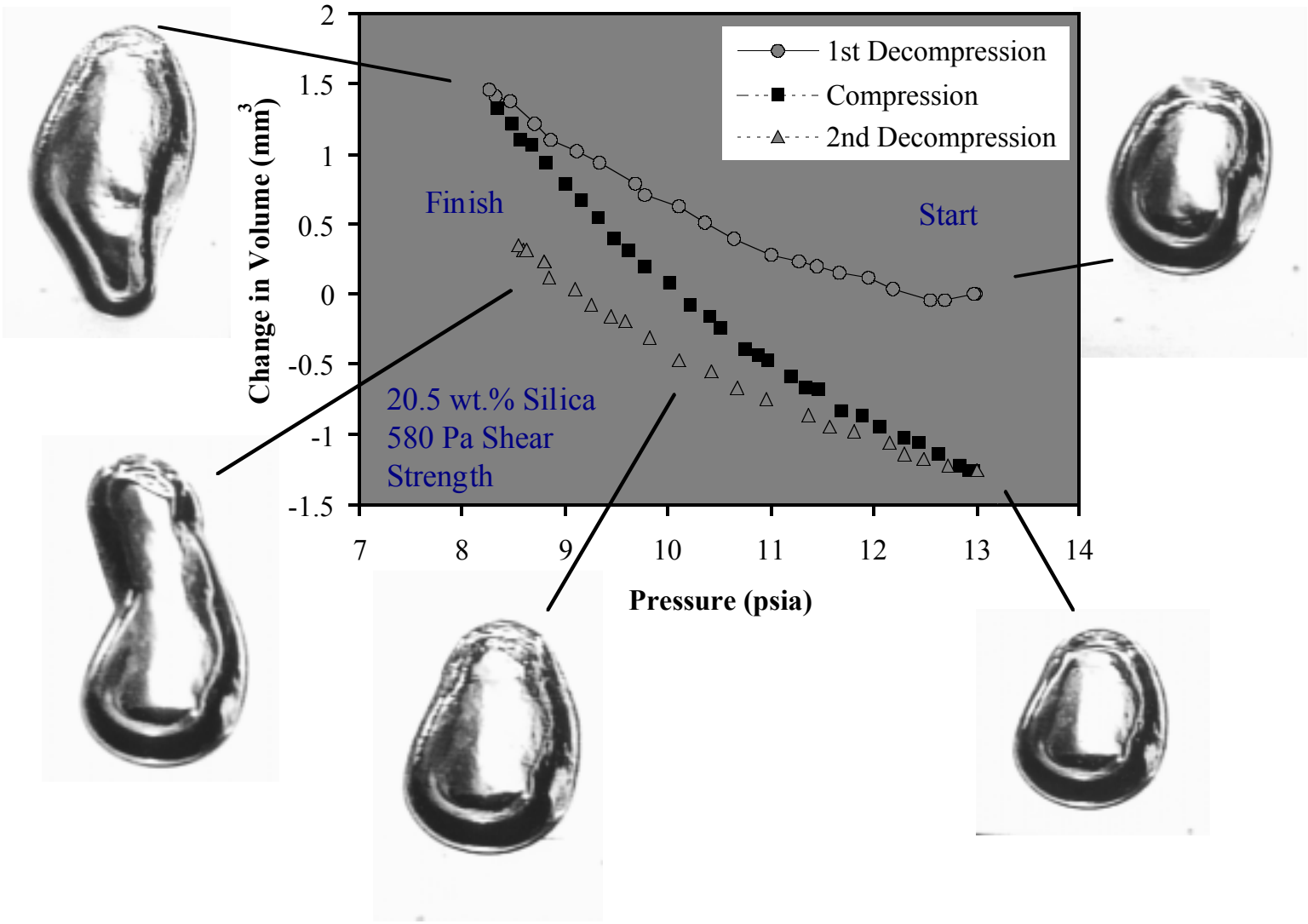
Silica particles and oils with matching refractive indices

- Slurry behavior appears representative of actual high-level tank waste
- Optical clarity is acceptable, but not excellent
- Range of achievable strengths smaller than desired

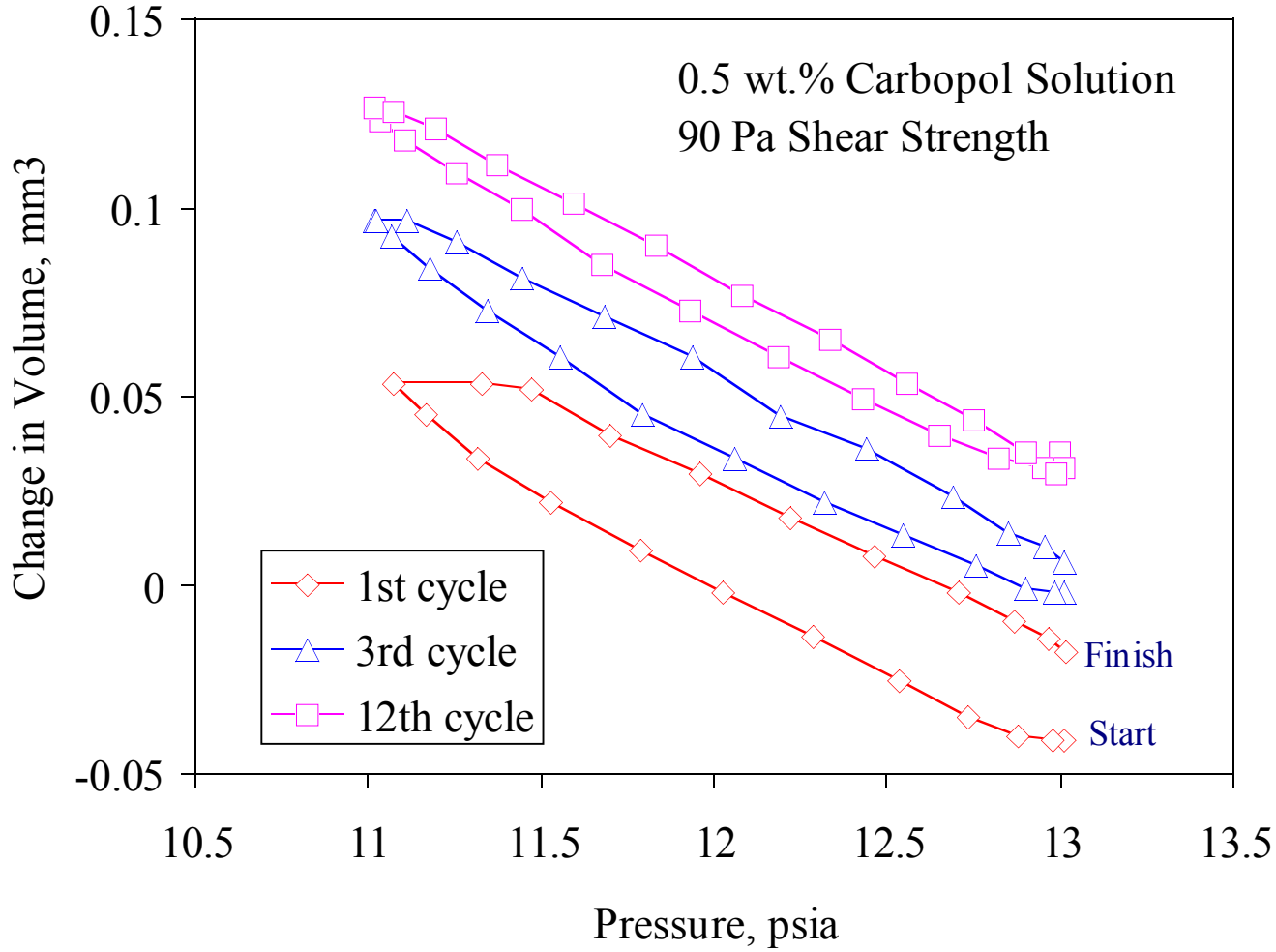
Carbopol polymer solutions

- Wide range of strengths easily achieved
- Optical clarity is excellent
- Material appears more elastic than actual high-level tank waste

Bubble Behavior in Silica Slurry



Bubble Behavior in Carbopol Solution



Summary

- Initial progress has been made on bubble behavior in continuum and particulate materials
- Mathematically rigorous solid mechanics theory completed for single pressure cycle
  - shows plastic failure region around bubble
- Constricted tube model essentially complete for particulate materials
  - Effective compressibility of gas shows hysteresis with pressure increase or decrease. Effective compressibility is much higher during pressure decrease than during pressure increase because of impulsive jump(s) during pressure decrease.
- Fluid mechanics studies are considering Bingham fluids and the unusual dependence on boundary conditions
- Single-bubble experiments show interaction of bubbles with waste during pressure cycling

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