

Multiscale Modeling of Active Brazing

Michael Chandross

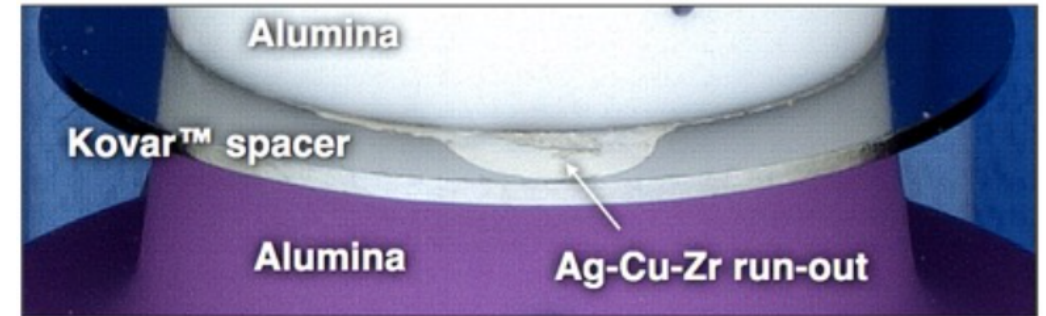
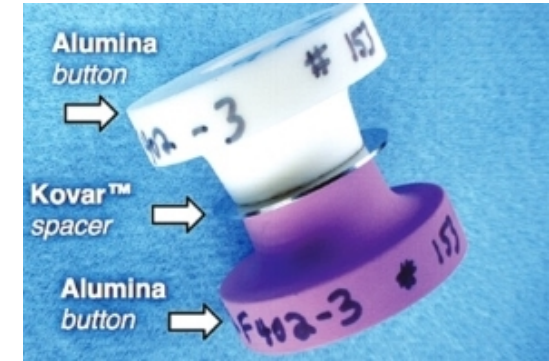
Ian Winter, Eric Rothchild, Jaideep Ray, Edward Arata, Ping Lu,
Jeffrey Horner, Scott Roberts, David Kemmenoe, Anthony
McMaster and Anne Grillet

World Congress on Computational Mechanics, Vancouver, BC Canada

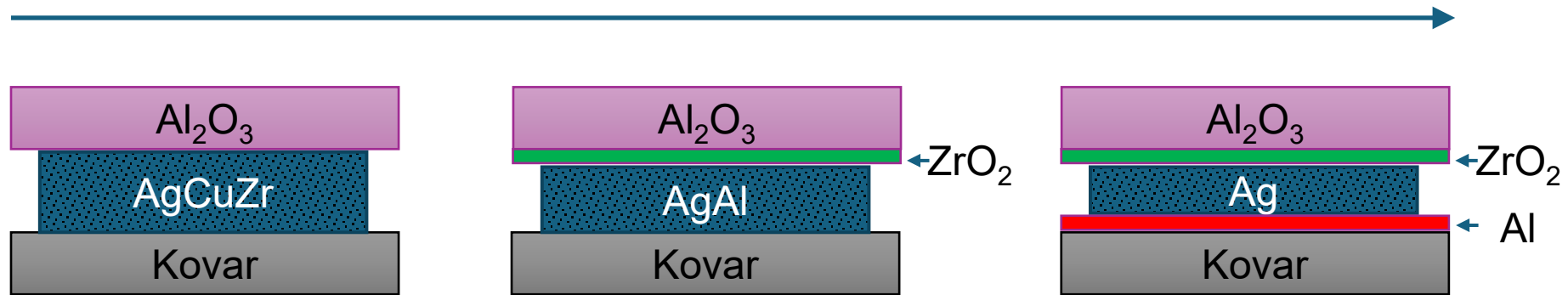
July 23, 2024

Introduction: Active Brazing

- Brazing
 - Method of joining materials
 - Braze alloy melts to join parts
- Active Brazing
 - Ceramic/metal joining (alumina to Kovar™)
 - Redox reaction aids wetting of ceramic
 - $4 \text{Al}_2\text{O}_3 + 3 \text{Zr} \rightleftharpoons 3 \text{ZrO}_2 + 4 \text{Al}$
- Braze run-out causes parts to be scrapped
 - Physical origins unclear
 - Possible wetting transition
 - Likely brought on by Al in braze alloy

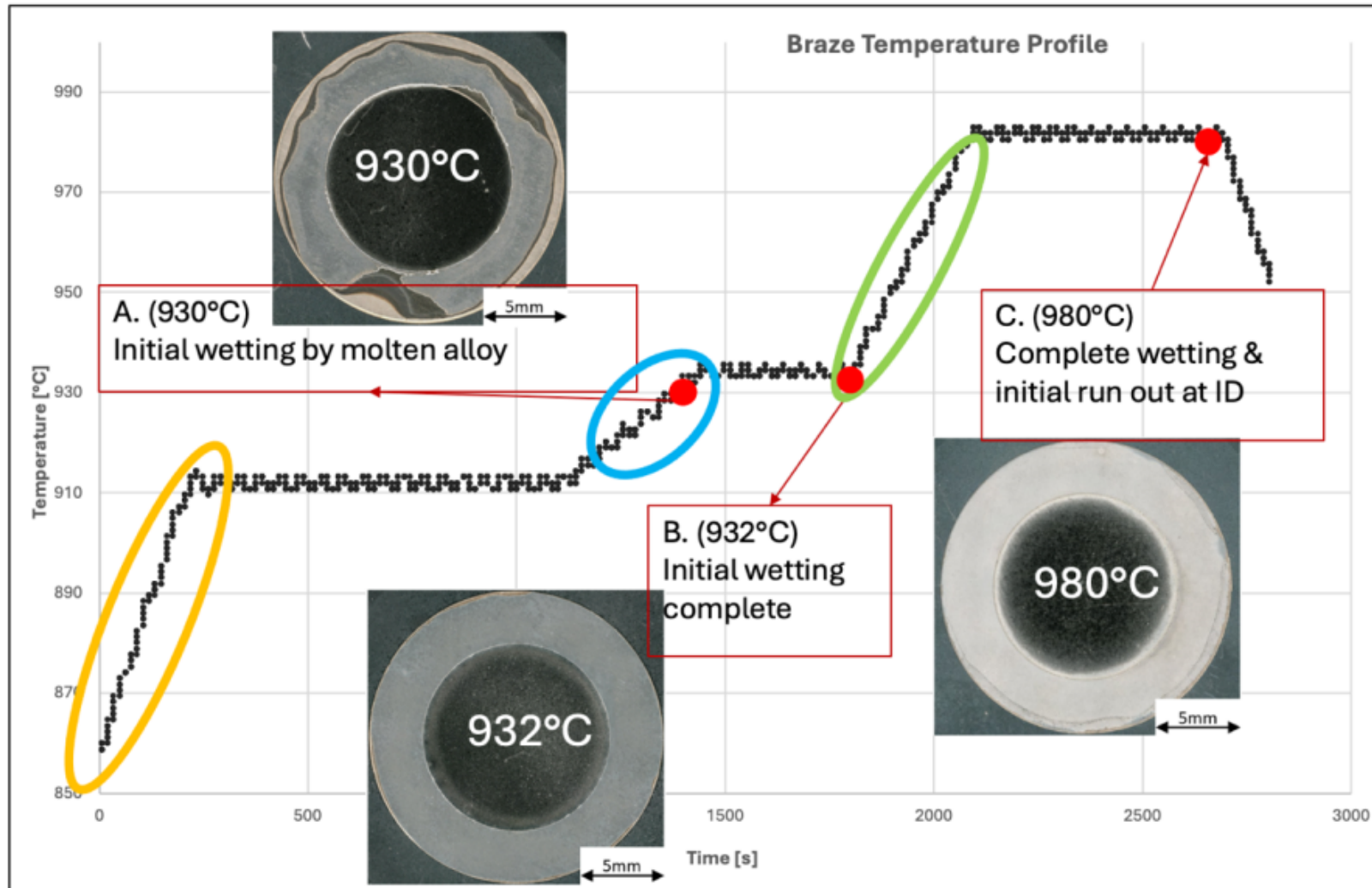


Schematic of the Process



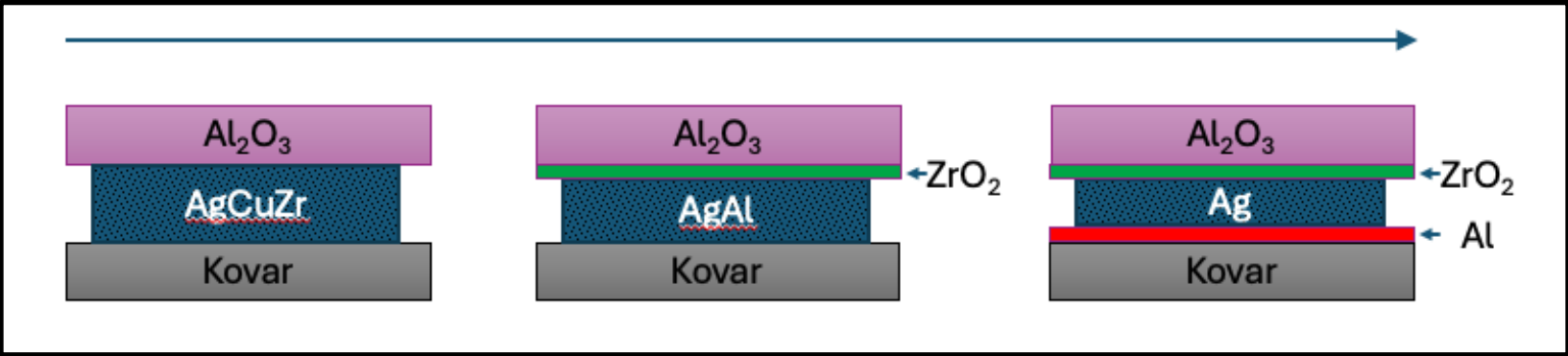
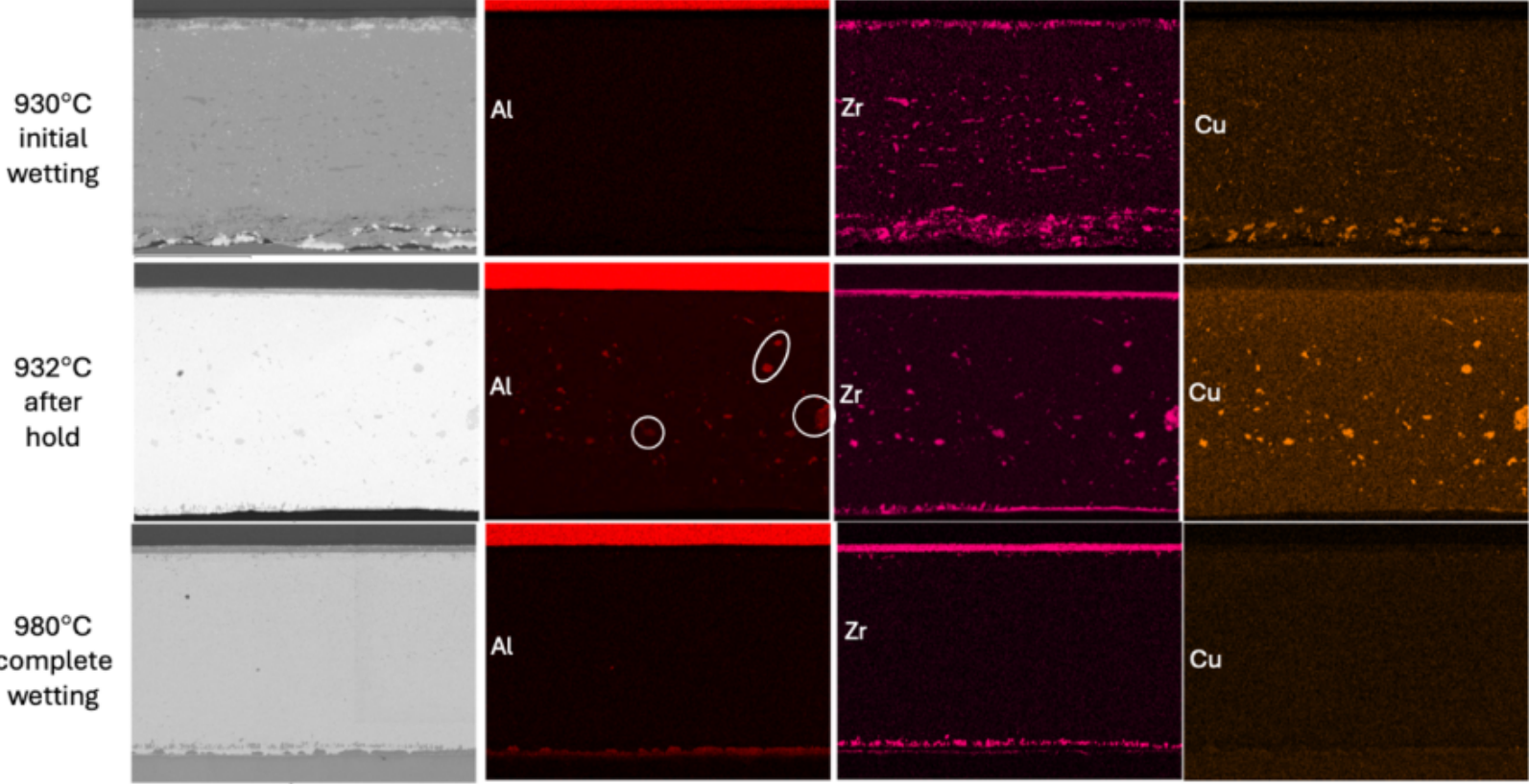
1. Braze joint (diamondite + KovarTM with braze alloy) heated
2. AgCuZr melts
3. Zr scavenges oxygen from Al_2O_3
 1. Forms ZrO_2 (suboxides?)
 2. Liberates Al
4. Al diffuses to KovarTM and forms surface layer
5. AgAlCu wets both surfaces
6. Occasionally runout!

Ramp/Hold Temperature Profile



Actual temperature profile is more complicated

EDS During Process



Goal of this Work

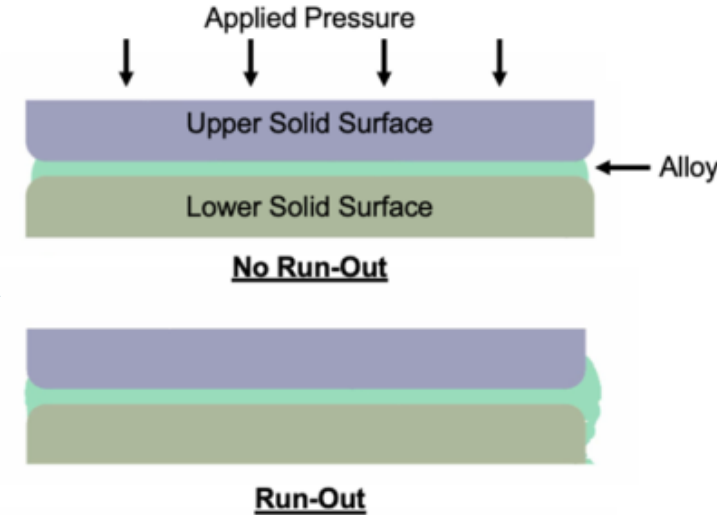
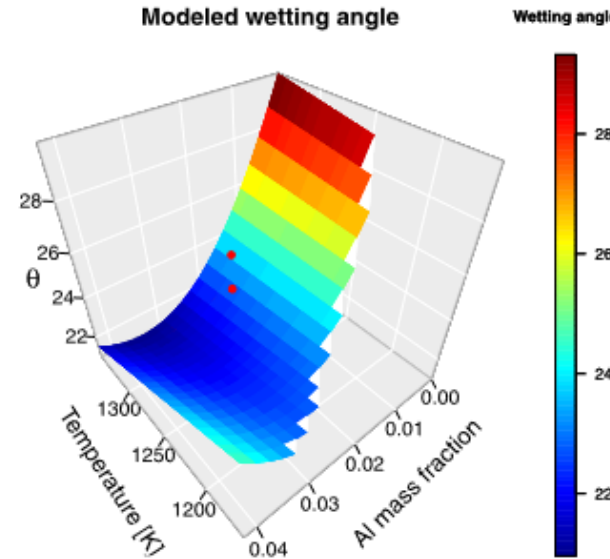
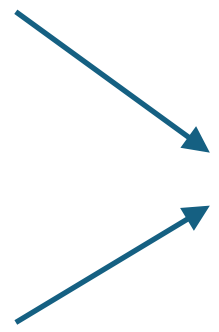
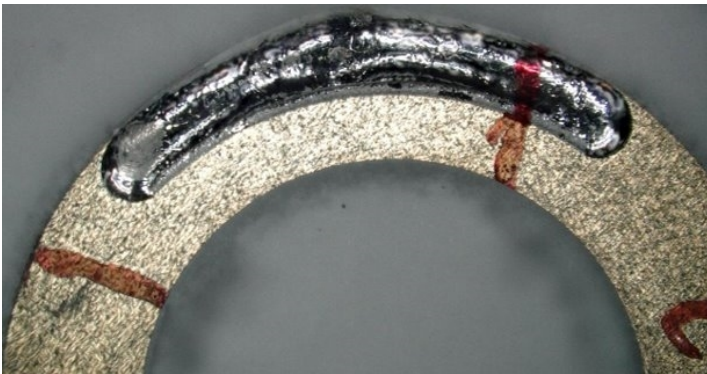
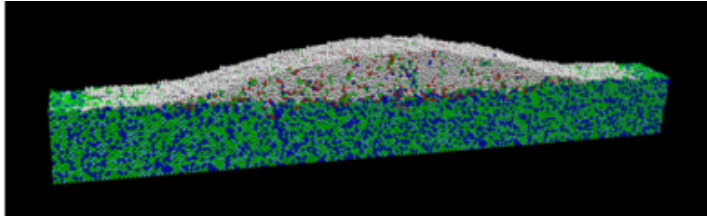
Chemically accurate
molecular dynamics
and experiments



Machine learning
generated constitutive
models



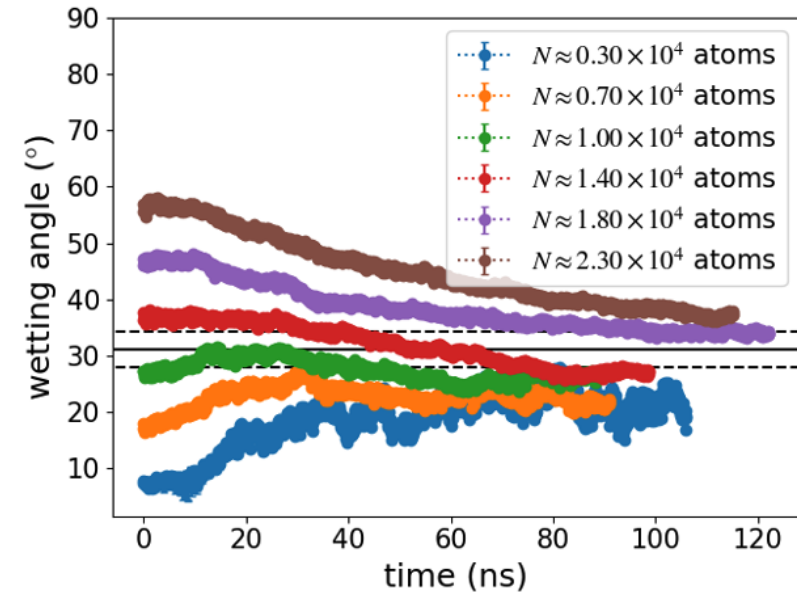
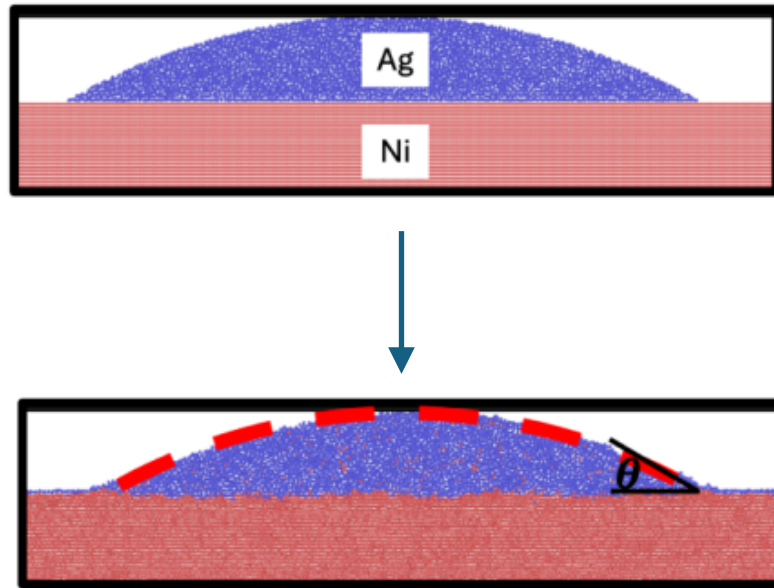
Engineering-scale
modeling of the entire
brazing process



- Requires accurate models of:
 - diffusion
 - wetting
 - Surface energies of substrates
 - Surface tension of braze alloy
 - Interfacial energies
 - chemistry (rate constants)

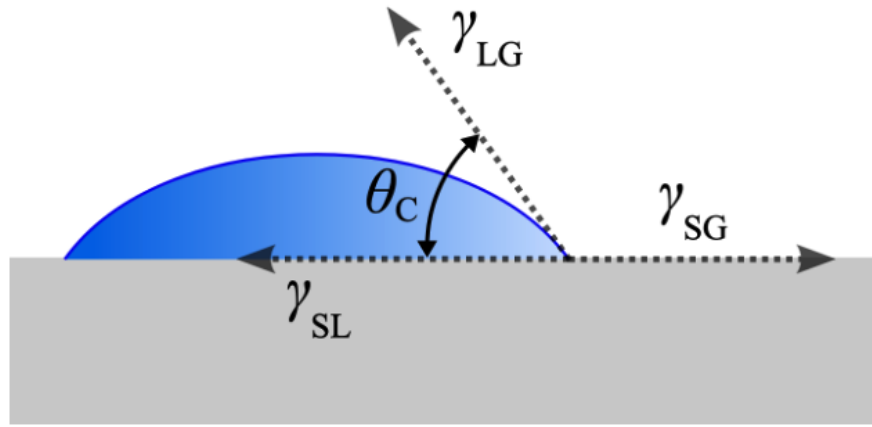
- Previous work
 - Metals only
 - Inaccurate results (alloying)
- Develop new potential with Penn State
 - metals & ceramics
 - chemistry

Molecular Dynamics Simulations (wetting and more)



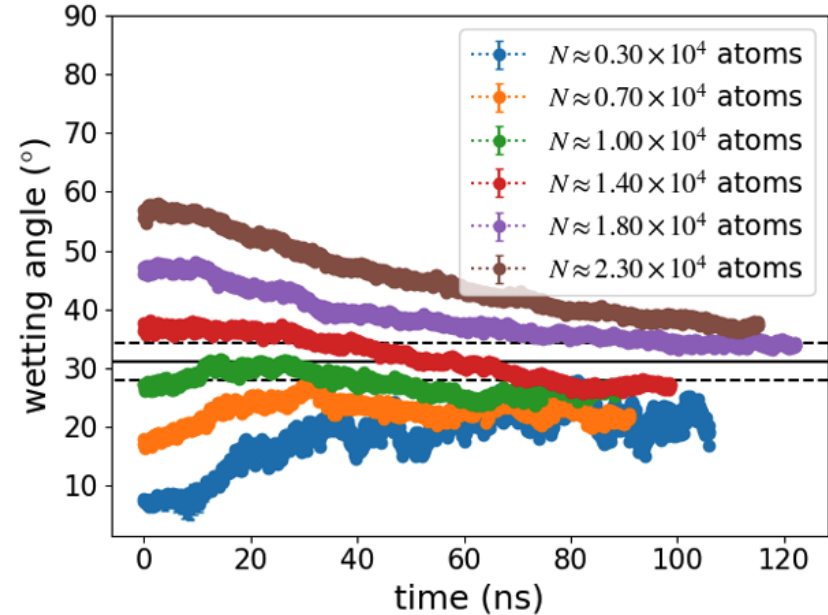
- Simulations of wetting are expensive
- Metal with ceramic requires reactive potential.
- Developed new reaxFF potential (Ag, Al, Fe, Ni, Co, O, Zr)
- Too slow to run for 100 ns
- Use Young's equation instead

Young's Equation for Wetting Angles



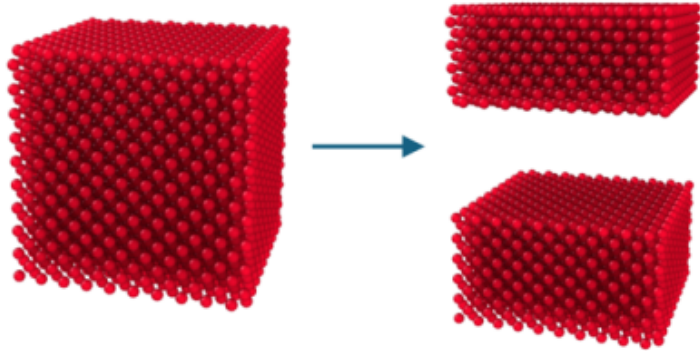
- Wetting is a balance of energies
 - Solid-vapor surface energy
 - Liquid surface tension
 - Solid-liquid interfacial energy
- Can calculate all of these with MD

$$\cos \theta = \frac{\gamma_{SL} - \gamma_{SV}}{\gamma_{LV}}$$



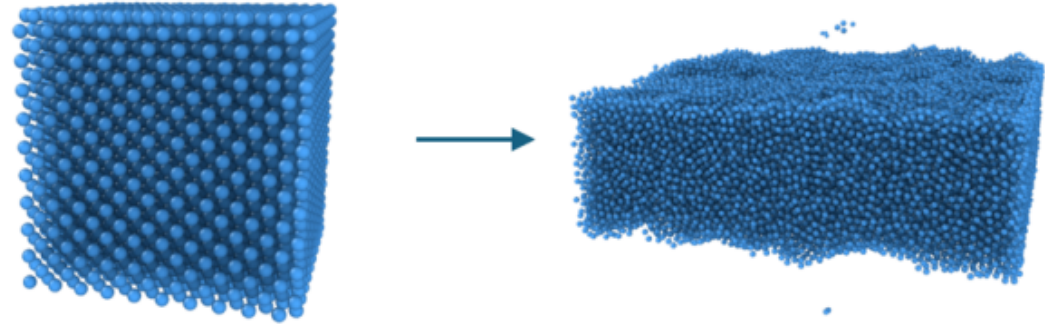
Calculating Energies with Molecular Dynamics

Solid surface energy γ_{SG}



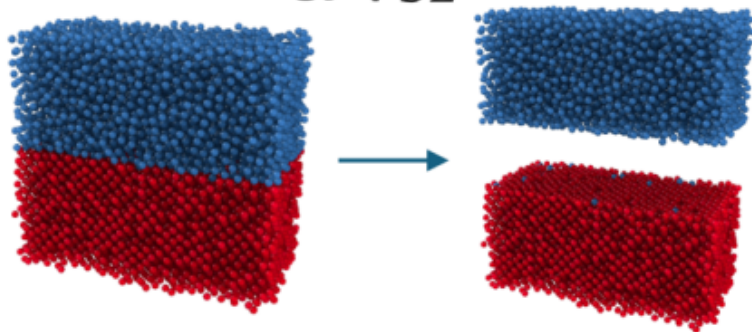
$$E_{bulk} = E_{separate} + 2 A \gamma_{SG}$$

Liquid surface tension γ_{LG}



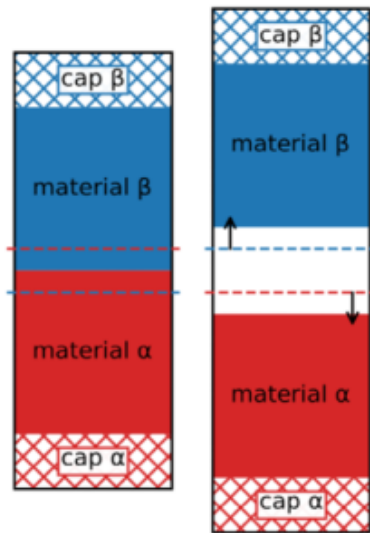
$$\gamma_{LG} = \int_{-\infty}^{\infty} \left[\sigma_{zz} - \frac{1}{2} (\sigma_{xx} + \sigma_{yy}) \right] dz$$

Interfacial energy γ_{SL}

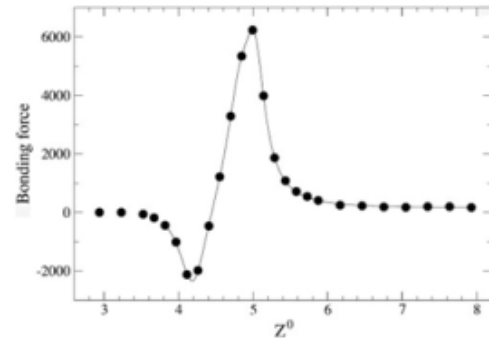


- Solid/liquid interfacial energy is difficult
- Use two methods
 - Double phantom wall
 - Capillary fluctuations

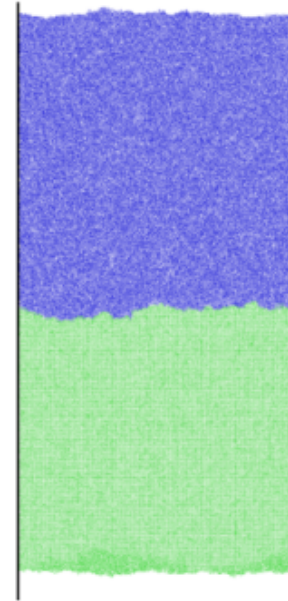
Interfacial Energies



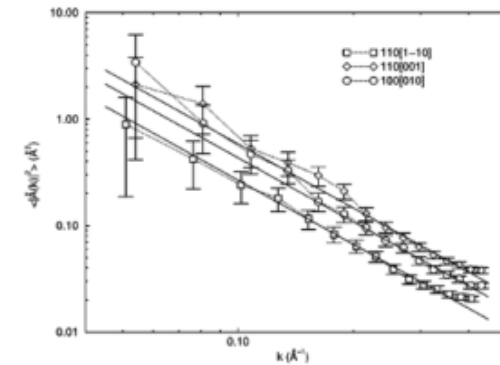
Double phantom wall: Push solid and liquid apart and measure forces



Leroy & Muller-Plathe, *J. Chem. Phys.* **133**, 044110 (2010)



Capillary fluctuations: Fourier transform of surface contours gives stiffness and interfacial energy



Hoyt, et al., *Phys. Rev. Lett.* **86**, 5530 (2001)

- Neither method objectively fast
- Both methods faster than wetting simulations!

Constitutive Equations

- Model forms for wetting angle (θ) and surface tension (σ)

$$\theta = w_0 + w_1 t + w_2 y + w_3 y^2 + w_4 y t$$

$$\Theta = \frac{\theta - \mu_\theta}{\sigma_\theta}; t = \frac{T - \mu_T}{\sigma_T}; y = \frac{Y_{Al} - \mu_Y}{\sigma_Y}$$

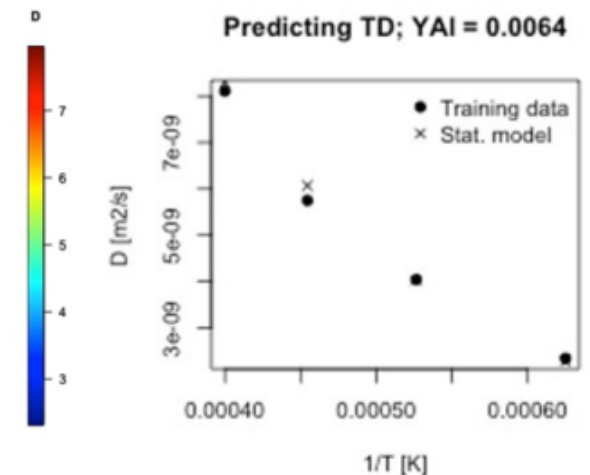
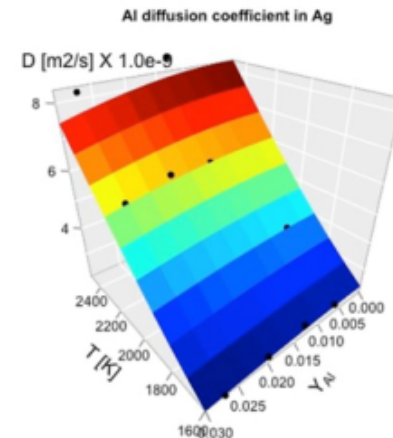
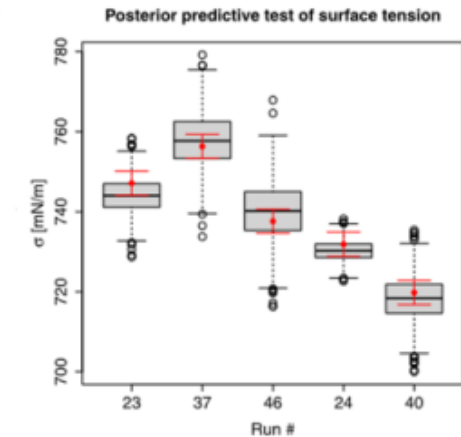
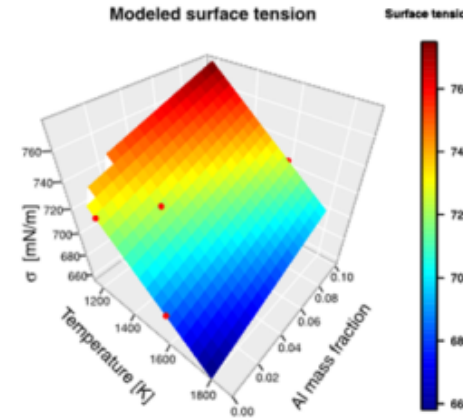
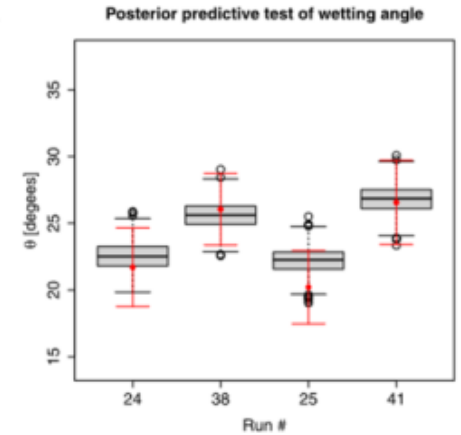
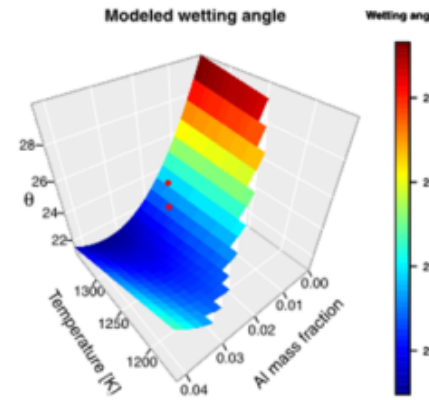
$$\sigma = aT + bY_{Al} + cT^2$$

- Several runs used for posterior predictive analysis
- Predictions showed good agreement

- Model form for diffusivity (Arrhenius): $D(Y, T) = D_0(Y) e^{T_0/T}$

$$T_0 = -5732, \ln(D_0) = -16.31 - 128.2 * Y_{Al}^2$$

- 128.2 is uncertain, p value = 0.05, $R^2 = 0.9895$



Finite Element Modeling Implementation

- To mechanically model this process, we use conformal decomposition finite element modeling (CDFEM) in combination with solid mechanics modeling to facilitate the fluid-structure interaction
- Alloy/atmosphere interface is captured using a level set (ϕ) approach

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

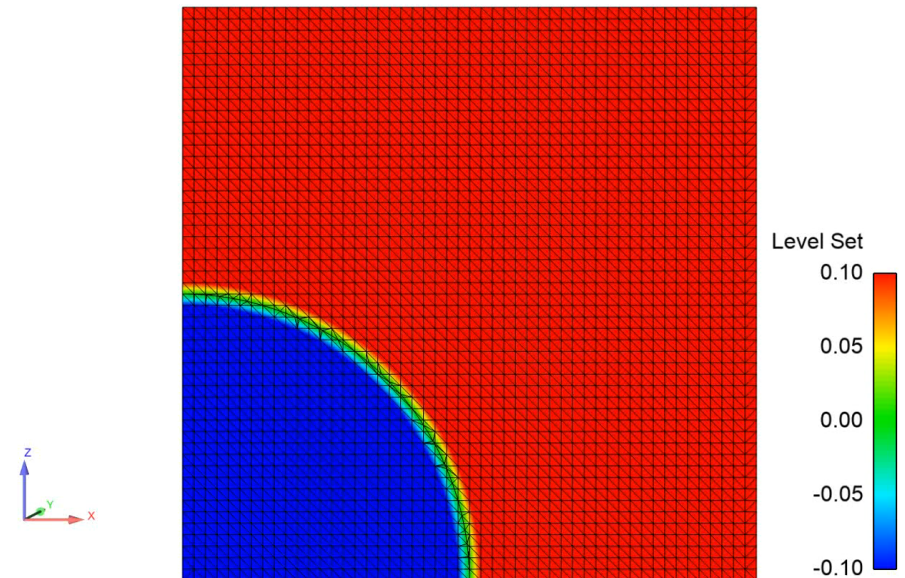
- A slip boundary condition is used in combination with a wetting angle (θ) condition to drive the wetting behavior of the alloy

$$\bar{t}_w \cdot \bar{T} \cdot \bar{n}_w = \frac{\mu\beta}{\Delta x} (\bar{v}_w - \bar{v}) \cdot \bar{t}_w$$

$$\bar{f}_\theta = \gamma(\bar{t}_w \cos \theta + \bar{n}_w \sin \theta)$$

- Surface tension (γ) is incorporated through an additional boundary condition on the alloy/atmosphere interface

$$\bar{n}_{int} \cdot (\bar{T}_l + \bar{T}_g) = -\gamma \bar{n}_{int} \nabla \cdot \bar{n}_{int}$$



Finite Element Modeling Implementation

- Simulation also involves fluid-structure interaction as the fluid is being “squeezed” between the two solid surfaces
- Solid mechanics are enabled through a separate arbitrary Lagrangian-Eulerian (ALE) mesh equation where the fluid Poisson’s ratio (ν_m) and Young’s modulus (E_m) are set close to zero

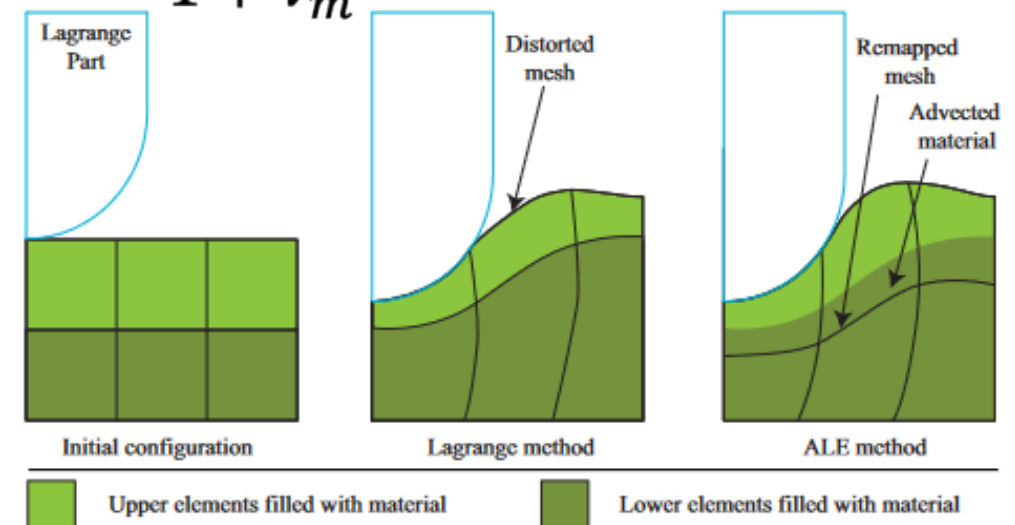
$$\nabla \cdot \bar{\bar{T}}_m = 0$$

$$\bar{\bar{\epsilon}}_m = \frac{1}{2} \left((\nabla \bar{d}_m)^T + \nabla \bar{d}_m \right)$$

$$\bar{\bar{T}}_m = \frac{\nu_m E_m}{(1 + \nu_m)(1 - 2\nu_m)} \text{tr}(\bar{\bar{\epsilon}}_m) \bar{\bar{I}} + \frac{E_m}{1 + \nu_m} \bar{\bar{\epsilon}}_m$$

- Solid mechanic motion is tied to fluid momentum through a boundary condition on the solid-fluid interface

$$\bar{n}_w \cdot \bar{v} = \bar{n}_w \cdot \frac{\partial \bar{d}_m}{\partial t}$$

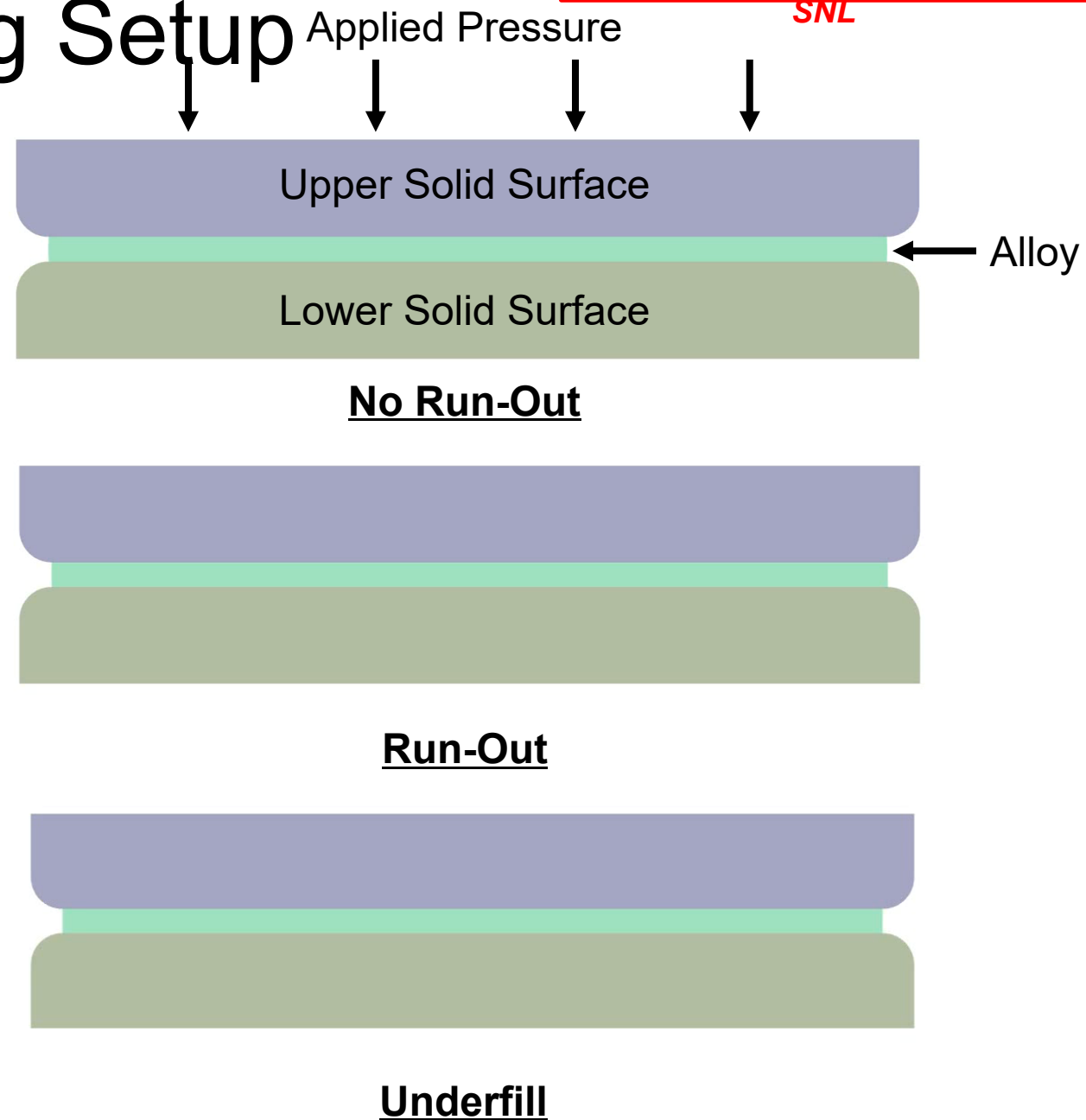


Finite Element Modeling Setup

slide courtesy of Jeff Horner,

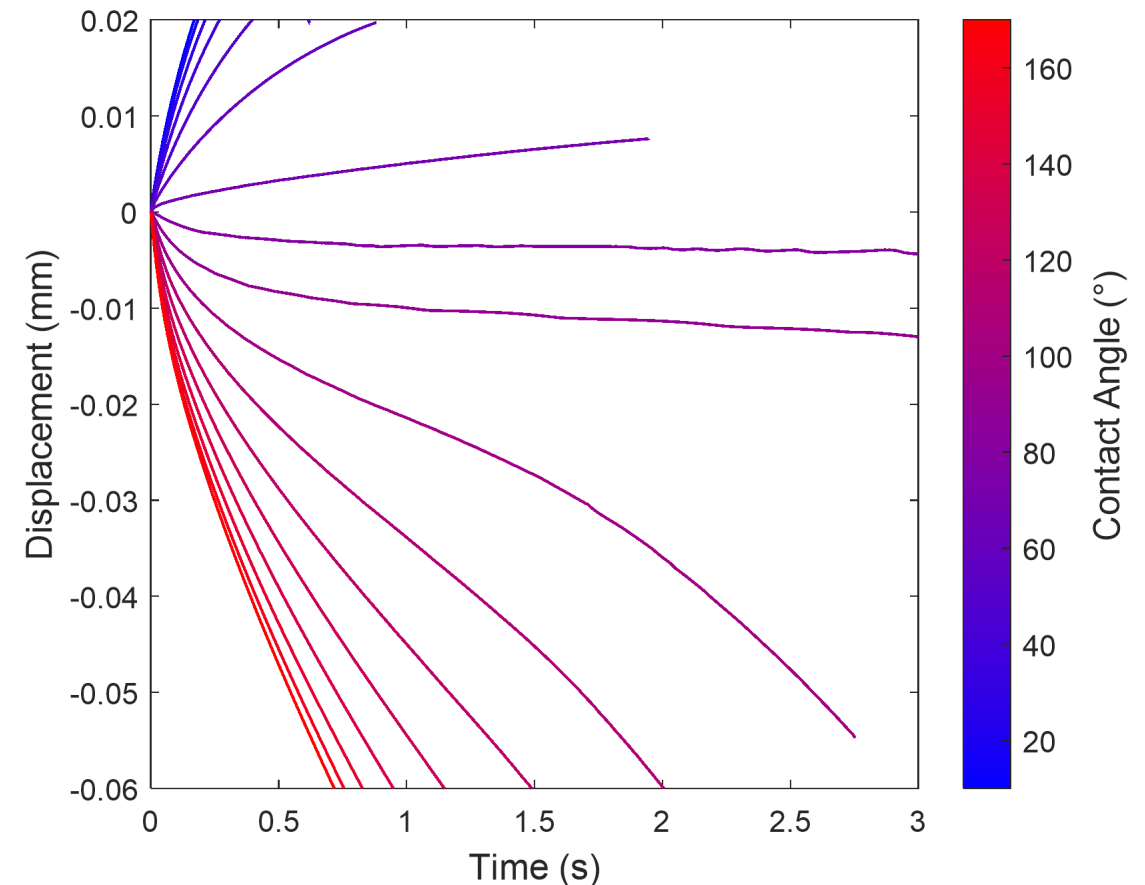
SNL

- 2D simulation with braze alloy “sandwiched”
- Pressure applied to top surface with fluid-structure interaction enabled
- Alloy flow dictated by contact angle and surface tension
- Al reacts at the ceramic surface and diffuses through the alloy to the Kovar, modifying the surface tension and wetting angle
- Zr within the alloy reacts preferentially at the ceramic surface forming an oxide that facilitates wetting
- Assuming first order reactions and Fickian diffusion for simplicity
- Can use FEM simulations to evaluate whether run-out will occur in experimental system



Quantifying Run-Out

- In the FEM simulations, run-out could be quantified through the displacement of the upper solid surface
- For cases without run-out, the displacement plateaus with respect to time
- Run-out cases exhibit an inflection point
- We approximate run-out to occur when the displacement of the upper surface decreases by at least 10%

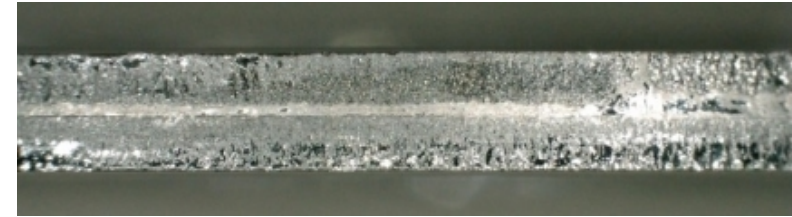


Model Validation – Comparison with Silver-Kovar

Experiments

- Experimentally measured contact angle for Silver on Kovar substrate
 - Values are temperature dependent
- Quantified run-out in a Kovar-Silver-Kovar stack using a run-out score which looks at 32 different sections of the edge to assess whether run-out occurred

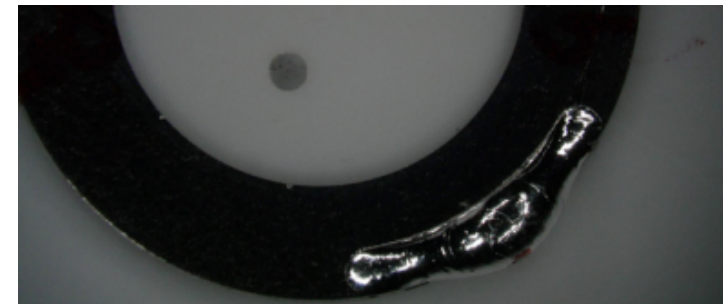
Minimal Run-Out; Run-Out Score = 0



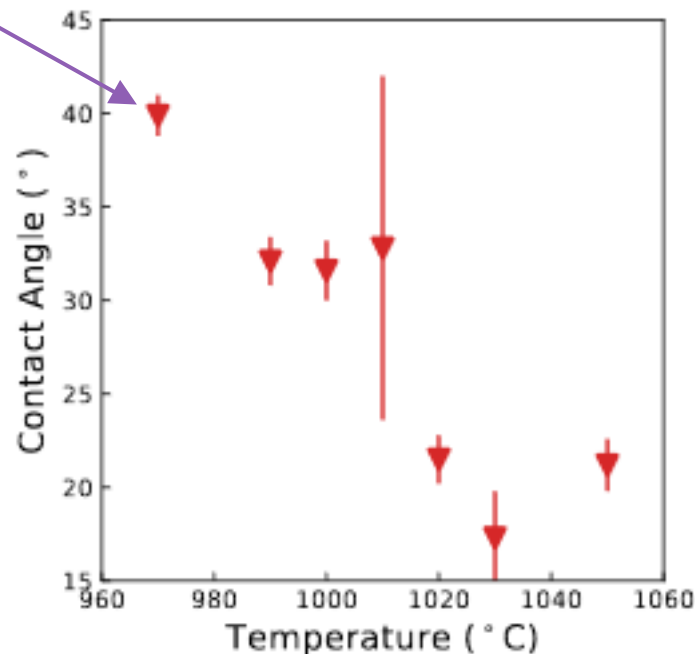
Significant Run-Out; Run-Out Score ~ 32



Blob Formation; Run-Out Score = 35

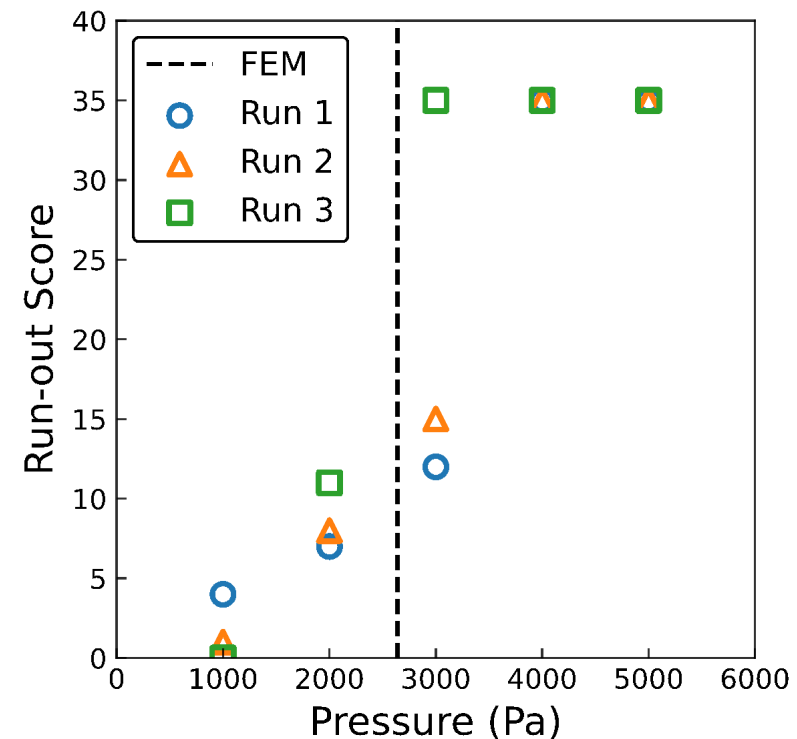
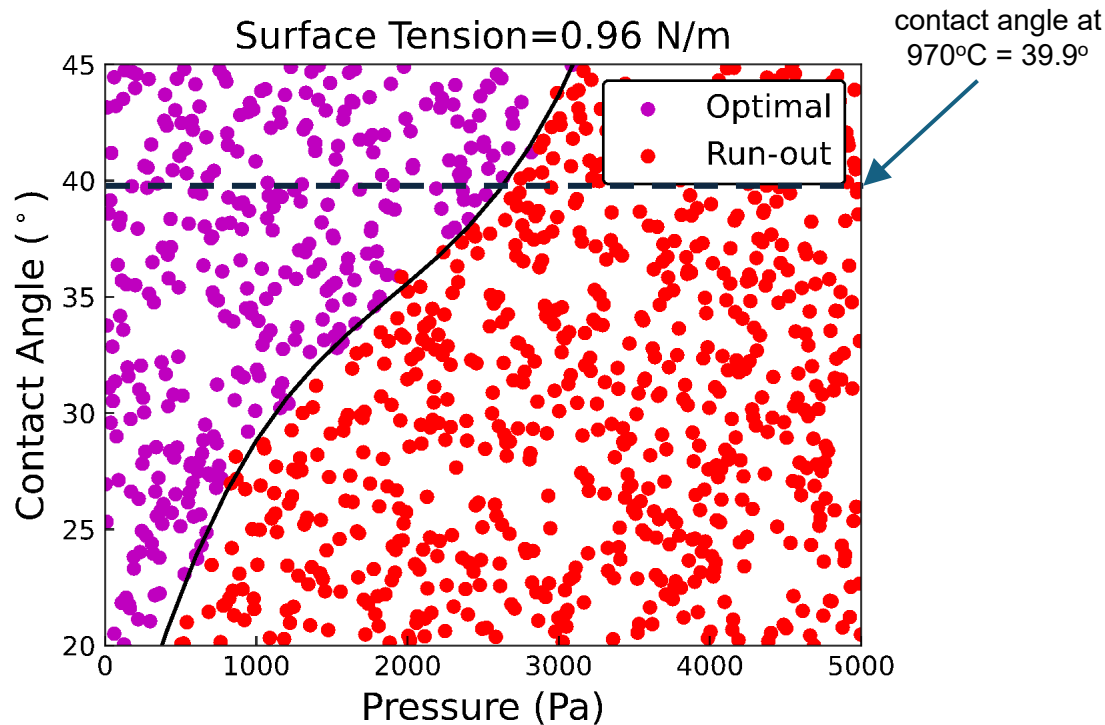


brazing at
T = 970°C
angle =
39.9°



Model Validation – Comparison with Silver-Kovar Experiments

- Compared model to experimental Kovar-Silver-Kovar stack tests with identical geometry
- Run-out pressures agree well with predicted values
- Experiments show little variation with temperature despite experimental contact angles varying



Summary and Conclusions

- Run-out is a consistent issue in active brazing
- Modeling successfully predicts runout
 - Begin with chemically accurate molecular dynamics
 - Use machine learning to develop constitutive models
 - Incorporate into FEA with moving fluid/solid boundary conditions
- Future work:
 - Theoretical treatment
 - Predicts drop stability
 - This system is on the edge of stability
 - Incorporate more reactions
 - Oxygen scavenging
 - Aluminide formation

