

Sandia Brazing Research and Modeling Capabilities*

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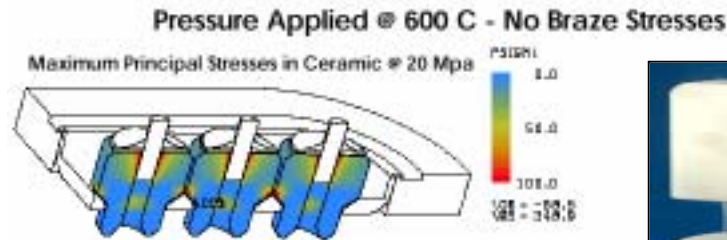


SECA SOFC Seal Meeting
July 8-9, 2003



Brazing is widely used to join metals and ceramics to each other for a variety of high reliability components

- headers
- connectors
- feedthroughs
- thermal batteries
- high voltage tubes
- electromechanical devices
- storage containers



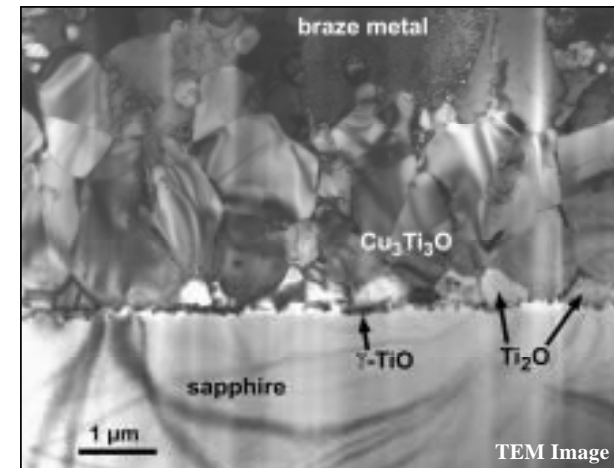
New active brazing alloys simplify hermetic joining issues

Basis: Oxide-forming additives to filler metal (e.g., V, Ti, Zr, Hf) promote direct wetting and adhesion to the ceramic materials (Al_2O_3 , Si_3N_4 , cermet, ...)

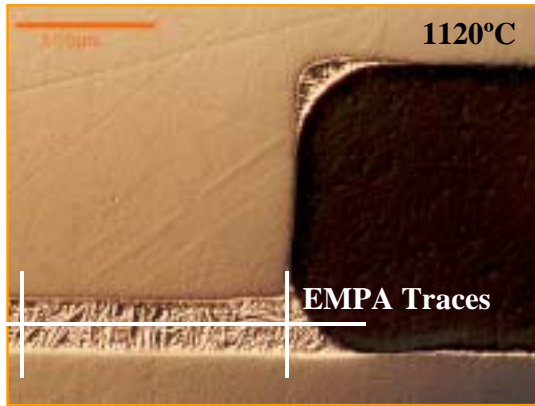
Payoff: Eliminate complex Mo-Mn metallize / Ni-plate & related processing for conventional alloys

Issue: Complex reactions between active braze elements and ceramic/metal base materials need to be understood to control and optimize process

TiO, Ti_2O & $\text{Cu}_3\text{Ti}_3\text{O}$ reaction products



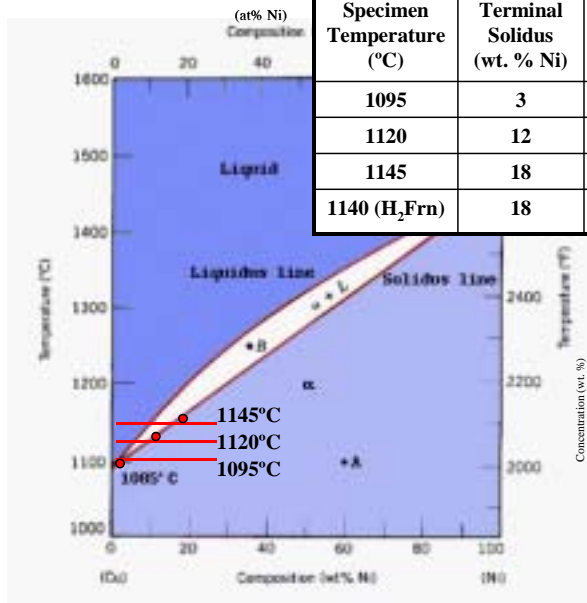
Braze flow visualization and dissolution reactions are important components in understanding the braze process



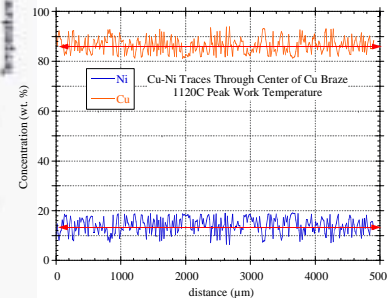
Cu-Ni binary braze flow & dissolution experiments



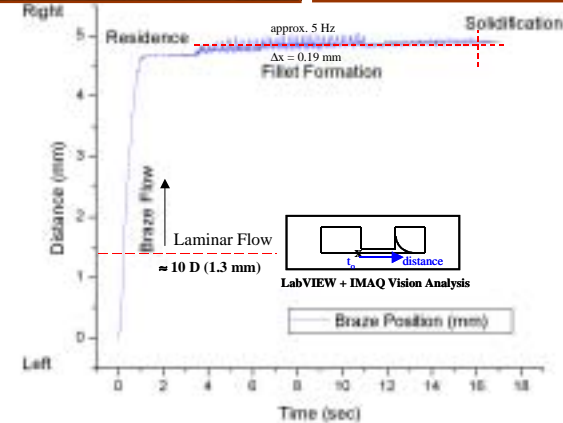
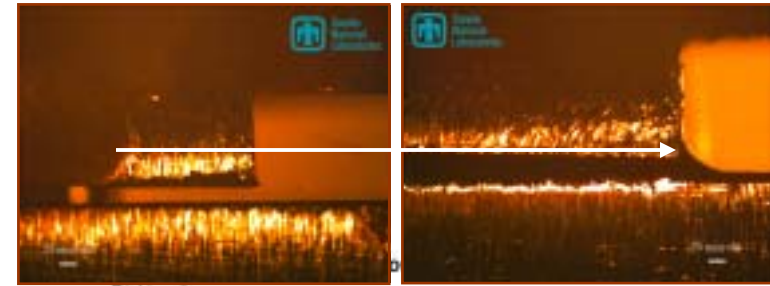
Specimen Temperature (°C)	Terminal Solidus (wt. % Ni)	EMPA Ni Content (wt. %)	Joint Thickness (μm)	Resident Time Above 1085°C
1095	3	6	175	0.5
1120	12	14	195	1.5
1145	18	20	210	3
1140 (H ₂ Frn)	18	10	190	660



Note: At 1145°C, $C_{Liquids} \sim 10$ wt. % Ni



Braze Capillary Flow Visualization

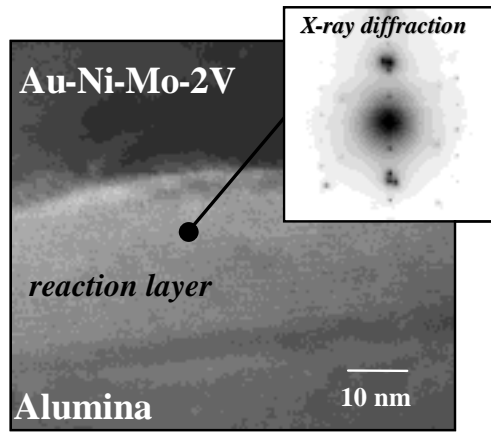


Recent Accomplishments

- Conference paper / presentations (IBSC'03; CSM Materials Science Seminar; C6 ASCI Solidification Working Group)
- *S&T of Welding & Joining* journal paper
- In-Situ Visualization of Braze Flow patent

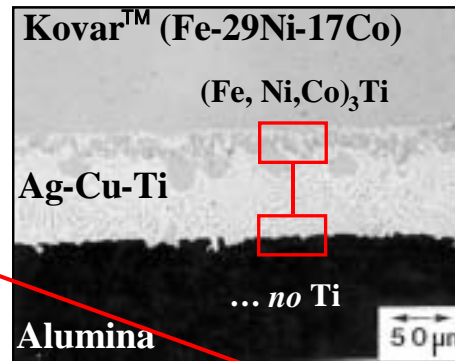
Active brazing technology is bridging a wide range of needs because of the fundamental knowledge being developed

Alumina ceramic / braze interface reactions vary significantly with active element

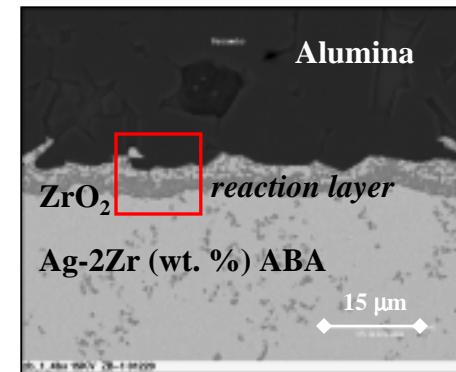


Competing reactions between dissimilar base materials can introduce unwanted results

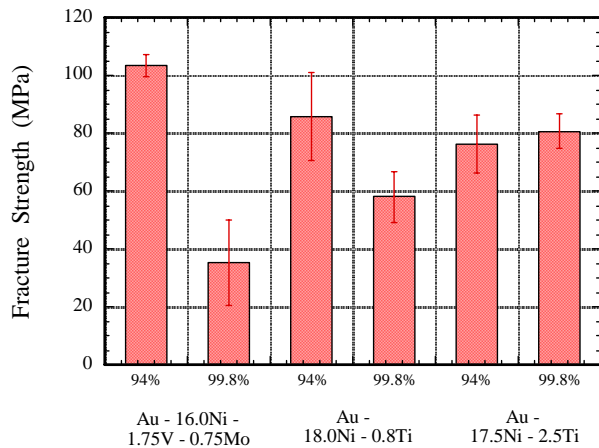
V activation



Ti activation
... compatibility issues



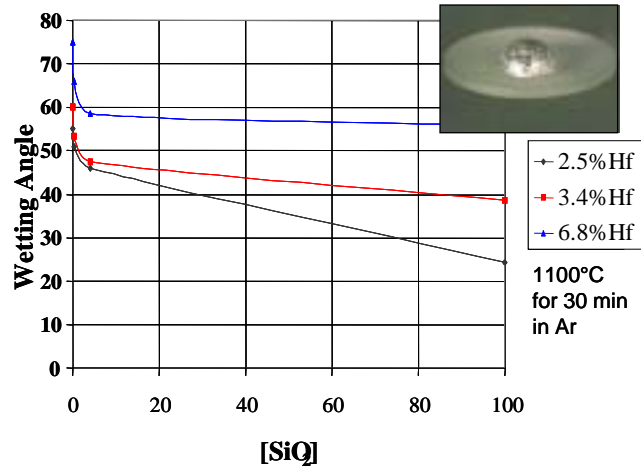
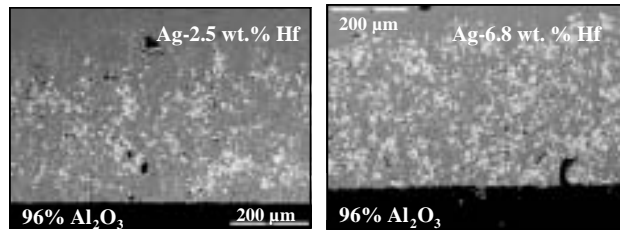
Zr activation



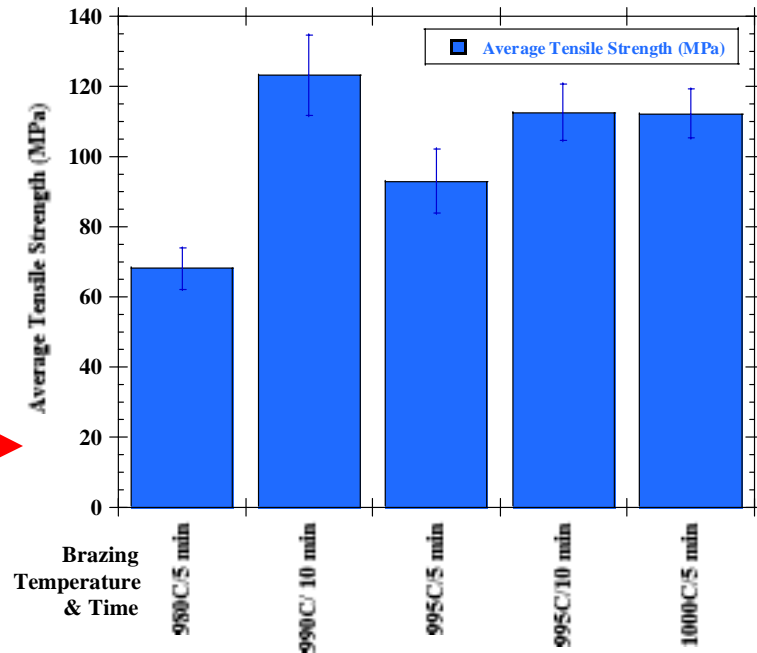
Fracture strengths for 94 & 99.8% alumina brazed with Ti & V-bearing Au-Ni filler metals exhibit dependency on glassy phase and more stable TiO_x reaction product

A multidisciplinary approach to characterizing the different brazing reactions and properties is necessary for success

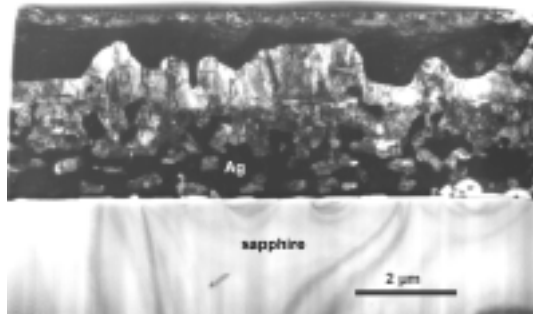
Determining effects of reaction products & microstructures on properties (hermeticity & strength) as a function of active element (thermodynamics / kinetics), base material (adhesion / dissolution), brazing conditions (temperature, time, atmosphere, heating cycle & gradients, orientation), surface preparation (ceramic air-firing).



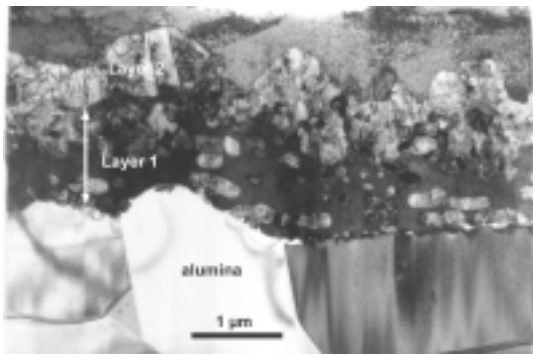
Al-500 alumina brazed with Ag-3.4Hf & 0.25 mm Kovar™



Ability to correlate braze interface reactions with desired properties will yield more producible & reliable joints

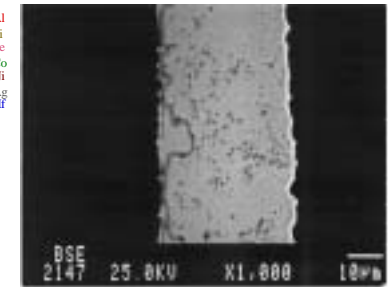
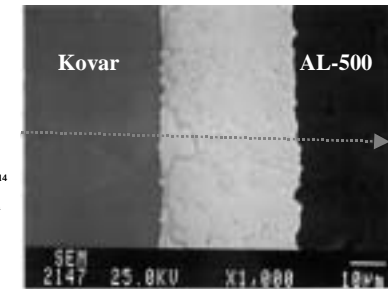
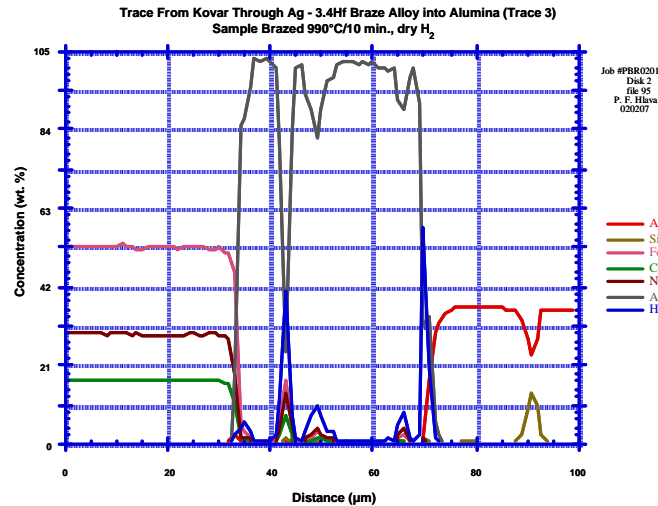


Single crystal sapphire & polycrystalline alumina brazed with Ag-2Zr



94% alumina brazed with Ag-2Zr is hermetic, fails in the bulk ceramic and has 130 MPa tensile strength

BSE image shows continuous HfO_2 reaction layer at ceramic interface



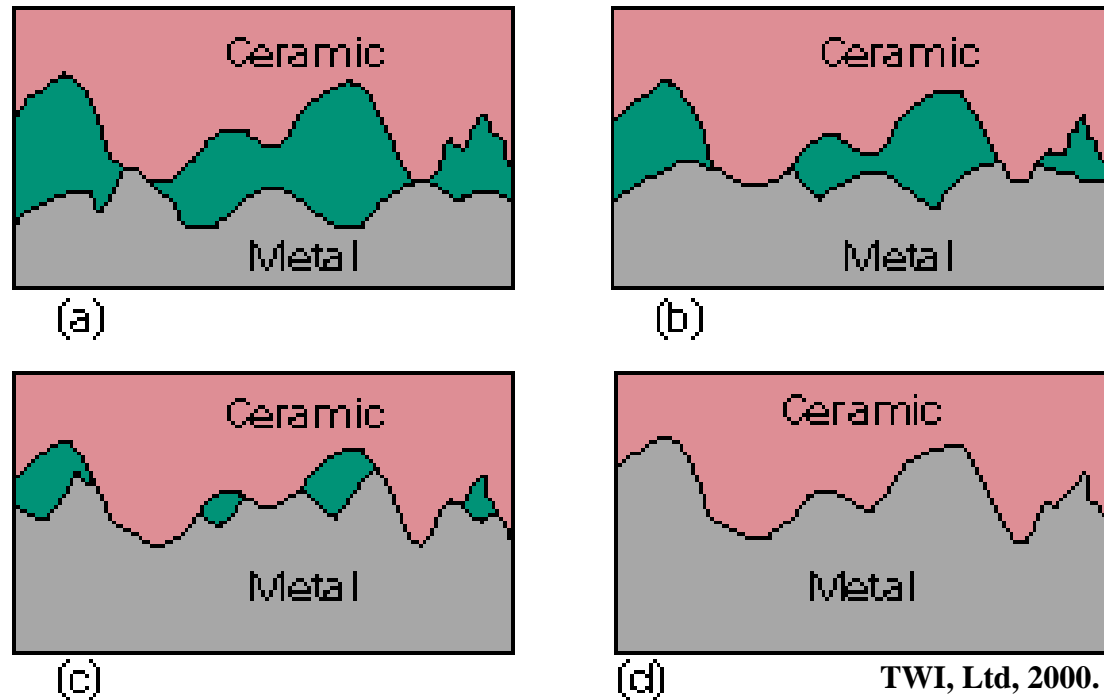
Ag-3.4Hf ceramic braze joints also demonstrate similar excellent properties (hermetic & 125 MPa)

Recent Accomplishments

- Implementation of active brazing in production
- Conference paper / presentations (ASM Materials Solutions 2002; AWS IBSC'03; ACerS 2003)
- *Metallurgical Transactions A* paper (in publication)
- Coating System for Direct Ceramic Brazing patents

Diffusion bonding offers an alternative joining process

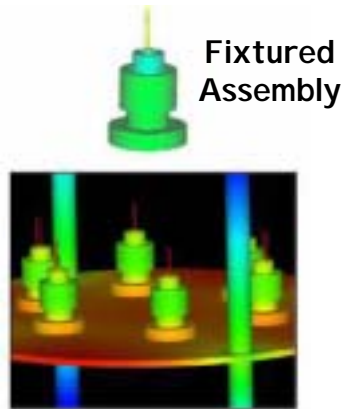
Ceramic-metal and metal-metal joints are possible



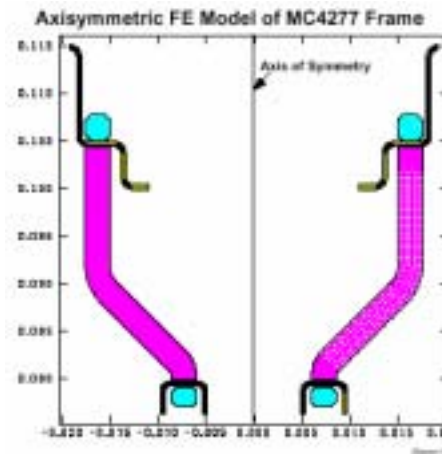
- a) Hard ceramic and comparatively soft metal surfaces come in contact (**T, t, P**)
- b) Metal surface yields under high local stresses
- c) Deformation continues mainly in the metal, leading to interface diffusion & void shrinkage
- d) Metallurgical bond is formed

FEA Brazing Model Development & Validation

Thermal Responses



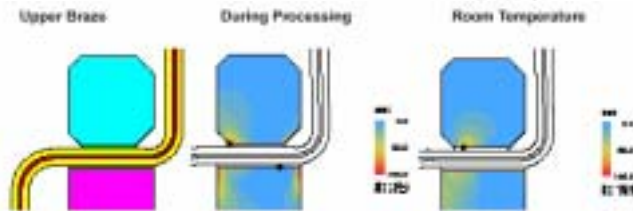
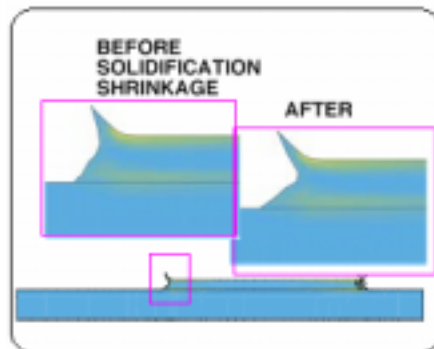
Stress & Failure Predictions



Design & Production

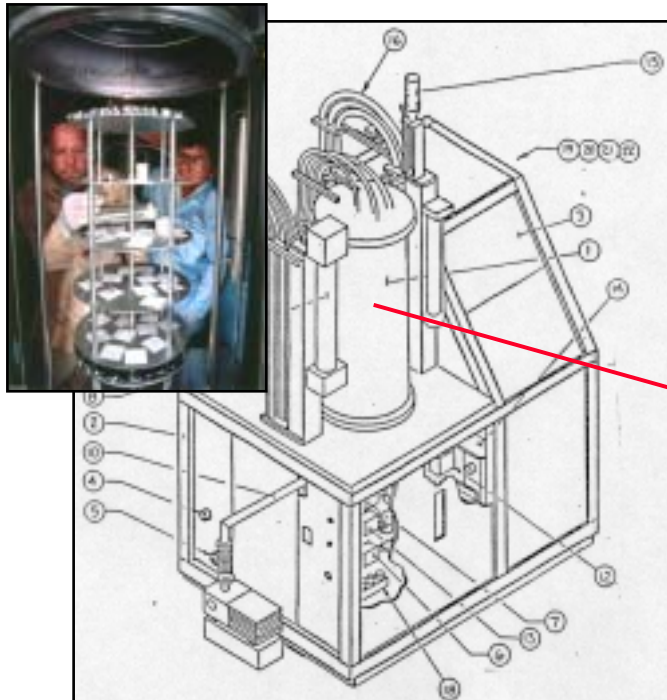


Fluid Flow Responses

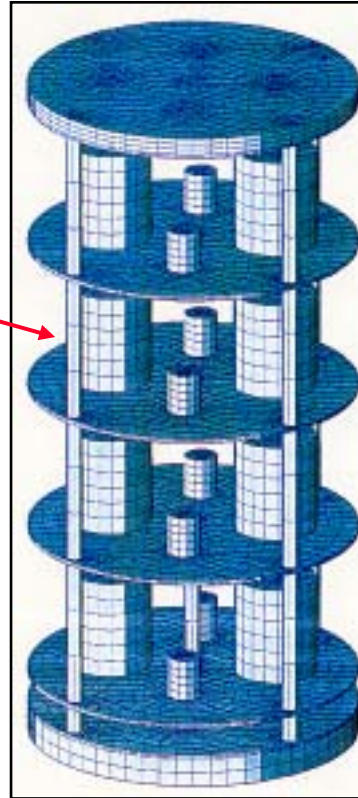


Engineering Sciences Center Simulations

FEA Thermal Modeling of Furnace Brazing



Work Rack & Parts



- Nonlinear, 3-D transient thermal finite element code
- Ability to mesh very fine details (large node and surface radiation enclosure)

Typical Model Inputs

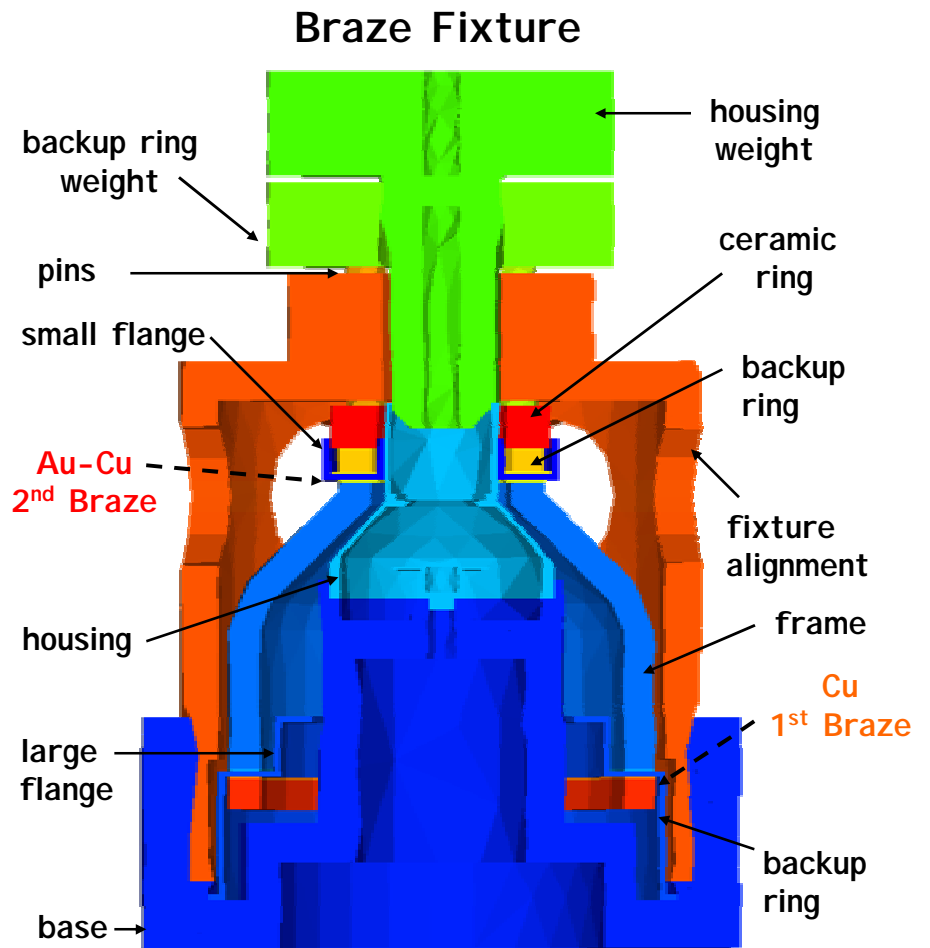
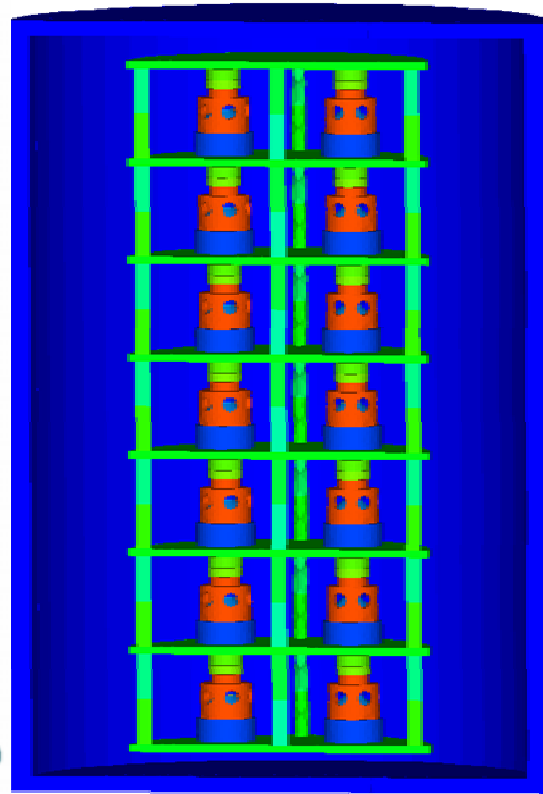
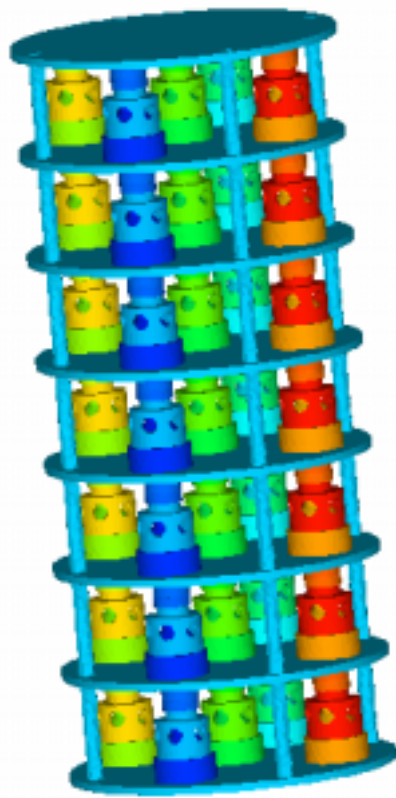
- materials density
- thermal conductivity
- specific heat
- emissivity
- thermal boundary conditions

- convective heating by hydrogen gas is assumed negligible
- heating driven by radiation from furnace elements & conduction from Mo shelves to work piece

Simulations processed on massively parallel teraflop compute server

Example: Two-Step, Cu + 50Au-50Cu Braze Assembly Process Characterization

FEA Thermal Model



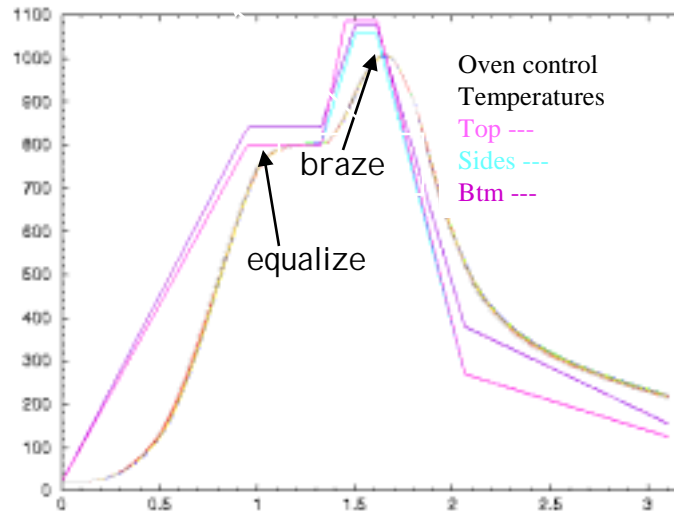
Process Space Simulations

Process Space:

- **Thermal boundary conditions:**
 - 1st ramp rate (10, 40°C/min)
 - 1st hold time (5, 25 min)
 - 1st hold temp (930, 960°C)
 - 2nd ramp rate (10, 40°C/min)
 - peak temp (1000, 1030°C)
 - time @ peak temp (0, 10 min)
 - cooling rate (10, 40°C/min)
- Number of shelves (4, 7)
- Number of fixtures/shelf (2, 5)
- Shelf type (solid, perforated)
- Fixture material (Mo, Kovar®)
- Emissivity (low, high)

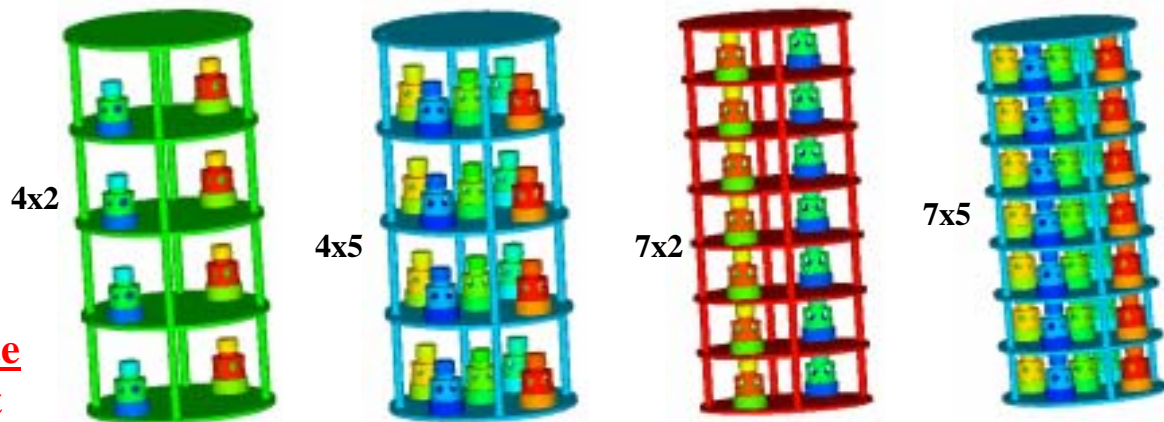
Data acquired:

- **Peak brazing temperature**
- **Time braze is melted**
- **Temperature uniformity**
- **Determine optimization parameters in process space versus experimental “point solutions”**



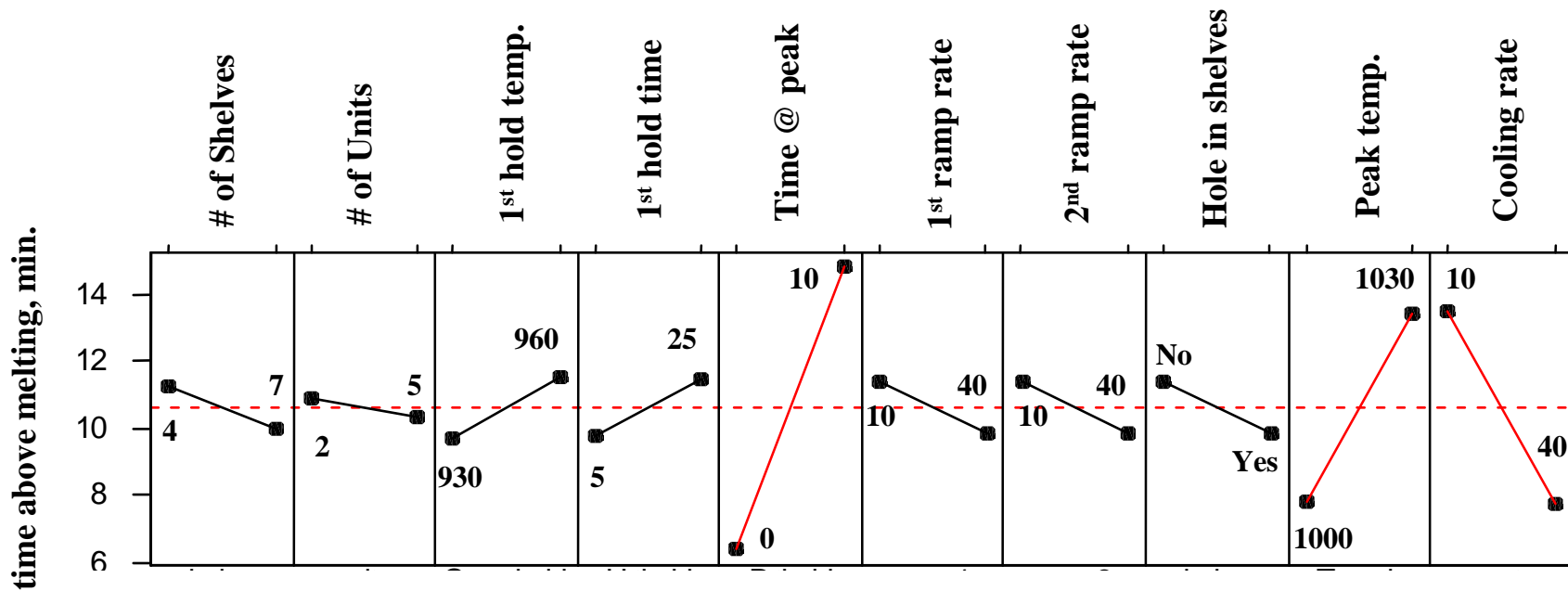
Trends at Peak Temperature:

- **Top and bottom shelves are hottest (trim heater settings)**
- **Middle shelf most uniform**
- **Center unit of five on each shelf is coolest (~ 5°C lower)**
- **10 min. hold at set temp. gives uniform heating, but increases time above melting**
- **Brazing temperature is uniform within individual units**



Two-level fractional factorial design for 12 factors in 16 unique trials

Example: DoE_x Screening Sensitivities for Thermal Characterization of Brazing Process



Factor sensitivities to the brazing conditions on the central shelf

— main effect factor significance