Bonding of Silicon Carbide Based Ceramics using Particulate Reinforced Ag-Cu-Ti Alloys

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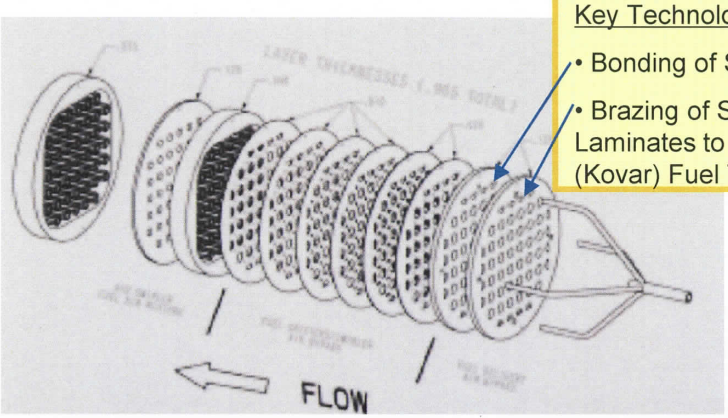
Outline

- **Background and Introduction**
  - Potential Applications
- **Experimental Details**
  - SiC Brazing using Ticusil
  - SiC Brazing using TiCusil + SiC particles
- **Characterization**
  - Optical Microscopy
  - Scanning Electron Microscopy (SEM)
  - Energy Dispersion Spectrometry (EDS)
  - Microhardness Measurements
- **Summary and Conclusions**

Need for Joining and Assembly Technologies

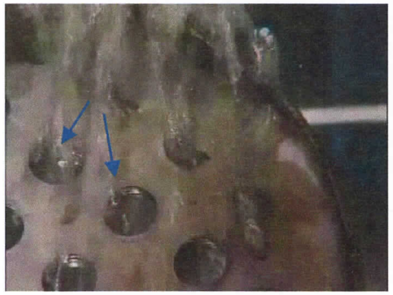
- **Joining is an enabling technology for utilization of advanced ceramics and composites in high temperature applications.**
  - **Aerospace Systems**
    - ***Aerospace and Space Propulsion Components (Combustor Liners, Exhaust Nozzles, Nozzle Ramps, Turbopump Blisks)***
  - **Non-Aerospace Systems**
    - ***Nuclear Industries, Land Based Power Generation, Process Industries, Heat Exchangers, Recuperators, Microelectronic Industries (Diffusion Furniture, Boats)***
- **The development of ceramic joining and assembly capability will allow the application of advanced ceramics and composites technology in a timely manner.**

Critical Need for Joining and Integration Technologies in Fabrication of MEMS Lean Direct Injector

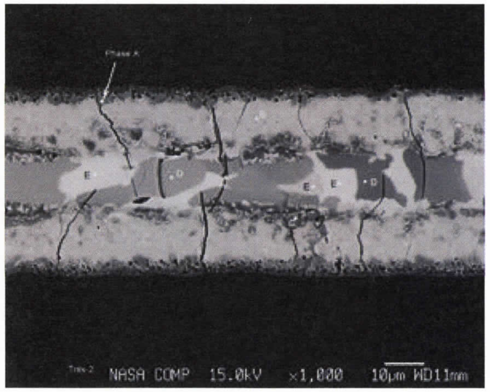


Key Technologies:

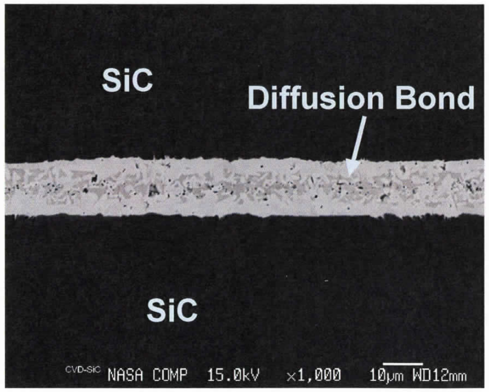
- Bonding of SIC to SiC
- Brazing of SiC Laminates to Metallic (Kovar) Fuel Tubes



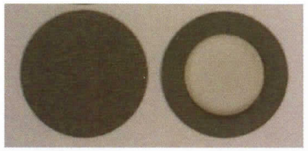
Previous joining approach using silicate glass resulting in plugged fuel holes and leaks in the SiC injector.

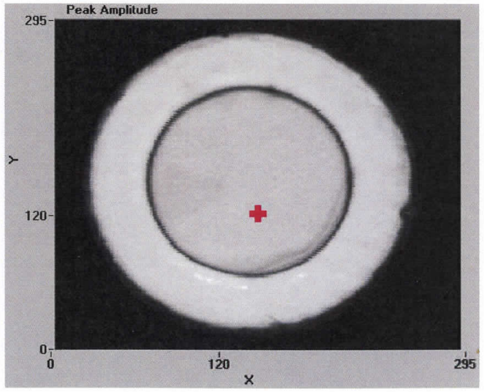


Un-optimized diffusion bond using a 38 micron thick alloyed Ti foil for an interlayer. Microcracks and non-uniformity are seen.



Optimized diffusion bond using a 10 micron thick PVD coating for an interlayer. A uniform bond with no microcracking is observed.





Above: Unbonded 1" SiC discs with a 0.65" diameter Ti coating on one of the discs. Below: Ultrasonic C-scan image of bonded discs.

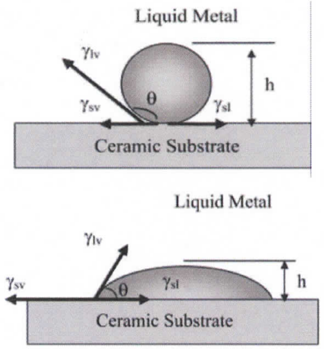
Technical Challenges in Bonding of Ceramics

- **Wetting of the Ceramic Substrate**
  - **Active Braze Alloys - Ti**

Contact Angle Between Ceramic and Liquid Filler Metal or Alloy

Ceramic Substrate	Filler Metal or Alloy	Temperature (°C)	Contact Angle (°)
SiC	Ag-28%Cu	930	160
SiC	Ag-35%Cu-1.5%Ti	850	10

Nascimento et al. Cerâmica 49 (2003) 178-198

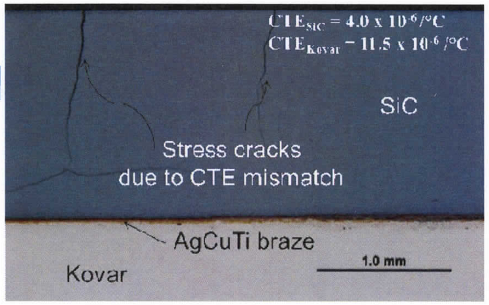


$$\cos\theta = \frac{\gamma_{sv} - \gamma_{sl}}{\gamma_{lv}}$$

- **Residual stress due to differing CTE of substrate and braze**
  - **Delamination and cracking in SiC substrate**
  - **Control CTE of braze by dispersing SiC particulate**

Technical Challenges in Bonding of Ceramics to Metals

- Residual stress due to differing CTE
  - Major problem in joining SiC to metal



  - Less severe in SiC-to-SiC joints but still important due to CTE mismatch between SiC and braze

- Control CTE of braze by dispersing SiC particulate

Background Information

- **C/C brazed to Ti-6Al-4V using SiC particles (4.6µm) in Ag-26.7Cu-4.6Ti**

  Qin and Yu, Joining of C/C composite to TC4 using SiC particle-reinforced brazing alloy. Mater. Charact., 61, 2010, 635~639.

- **C/SiC brazed to metals using (Ag-6Al)+Ti+C filler (as mixture of Ag, Al, Ti powders and short carbon fibers)**

  Wang et al, Reactive composite brazing of C_f-SiC composites to Ti alloy with (Ag-6Al)+Ti+C composite filler materials, Mater. Sci. Technol., 27(1), 2011, 49-52

- **C/SiC composite joined to Ti using Ag-Cu-Ti braze containing short carbon fibers**

  Lin et al, Joints of carbon fiber-reinforced SiC composites to Ti-alloy brazed by Ag-Cu-Ti short carbon fibers, J. Mater. Proc. Technol., 189(1-3), 2007, 256-261

Main Findings:

- Particles and fibers reacted with Ti, forming thin TiC_x layers
- Particles and fibers randomly distributed in braze
- Fibers preferentially aligned parallel to brazed interface

Materials and Key Properties

Materials:

- Substrate: CVD silicon carbide
- Braze matrix: Ticusil (68.8Ag-6.7Cu-4.5Ti, T_L: 900°C)
- Braze reinforcement: SiC particulates (size: ~20 μm)

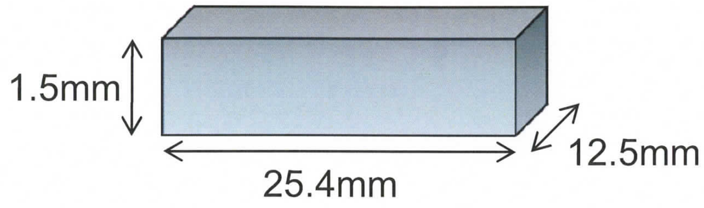
Generic Property Data

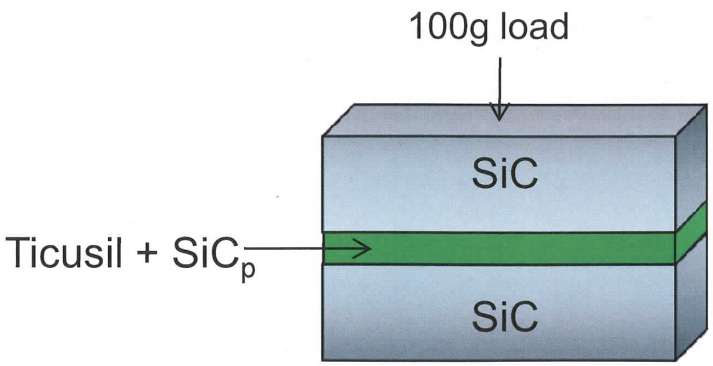
	E (GPa)	σ_y (MPa)	U.T.S. (MPa)	Thermal Conductivity (W/mK)	CTE ($x10^{-6}$ m/m*K)	Electrical Conductivity ($x10^{6}$ /Ωm)	% Elongation
Ticusil	85	292	339	219	18.5	29	28
SiC	466	-	-	300	4.0*	-	-

* 0 - 1000°C

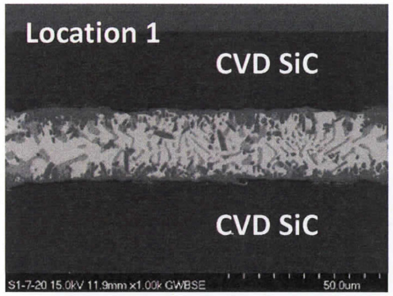
Experimental Procedures

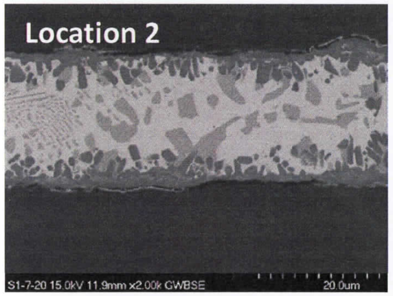
- Ultrasonic cleaning of SiC substrate in Acetone for 10 min
- Ticusil powder mixed with 5, 10 and 15 wt% SiC particulates (~20 µm) and glycerin to dough-like paste
- Paste applied using spatula and substrates sandwiched under 200g load
- Heated to 10°C above the braze liquidus temperature, isothermally held for 5 minutes
- Mounted in epoxy, polished and characterized

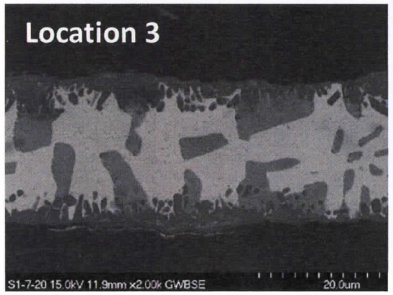


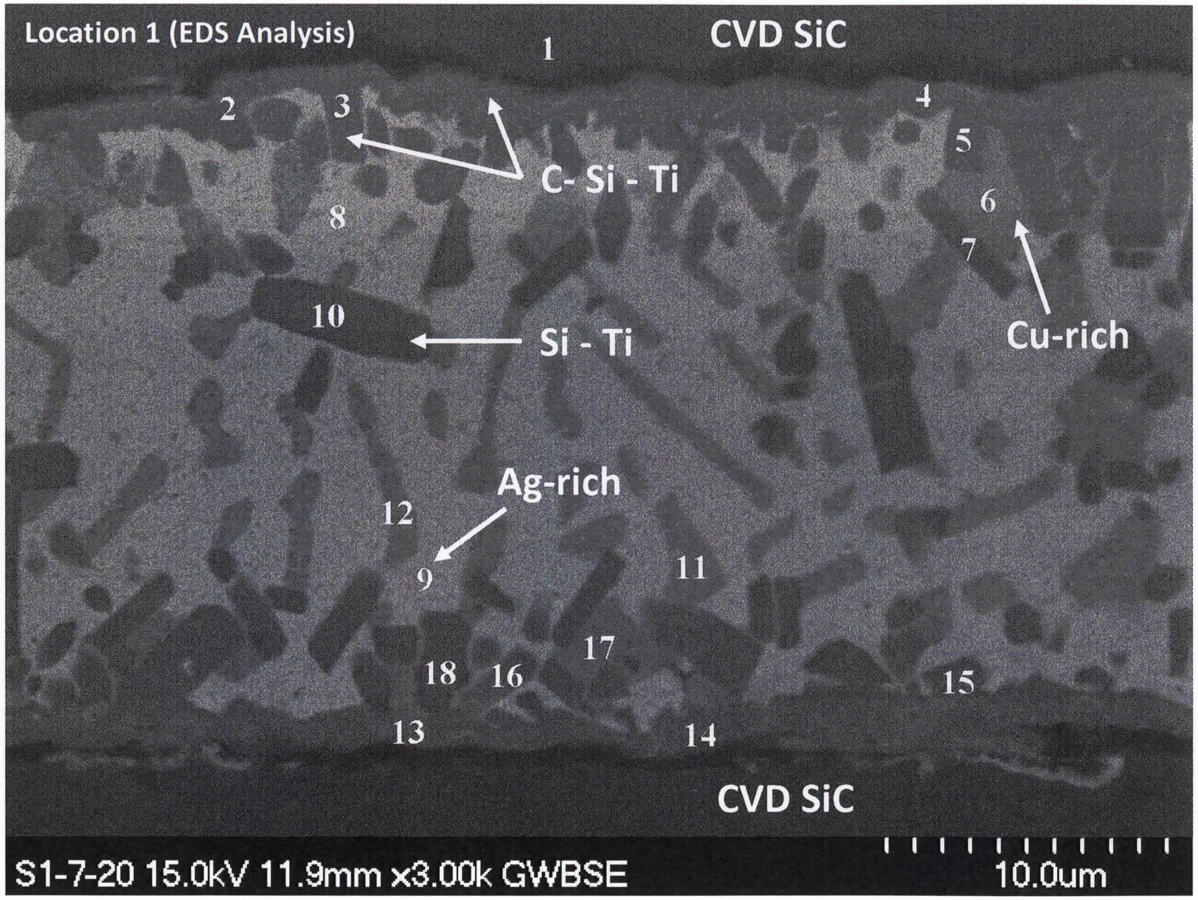


CVD SiC/Ticusil (0% SiCp)/CVD SiC

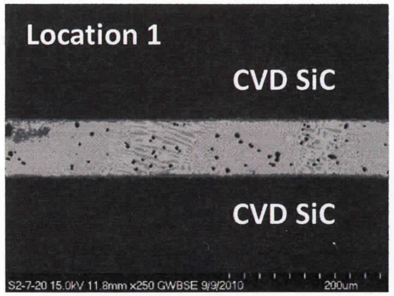


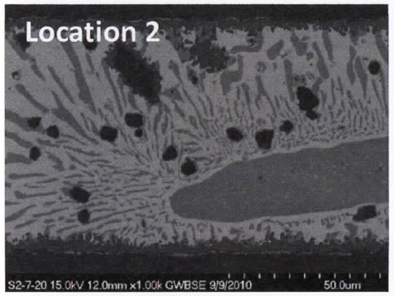


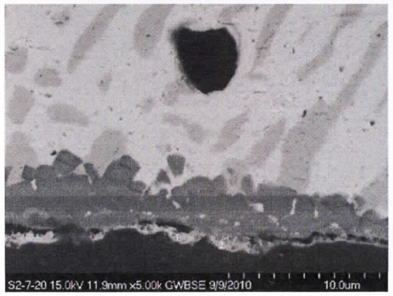


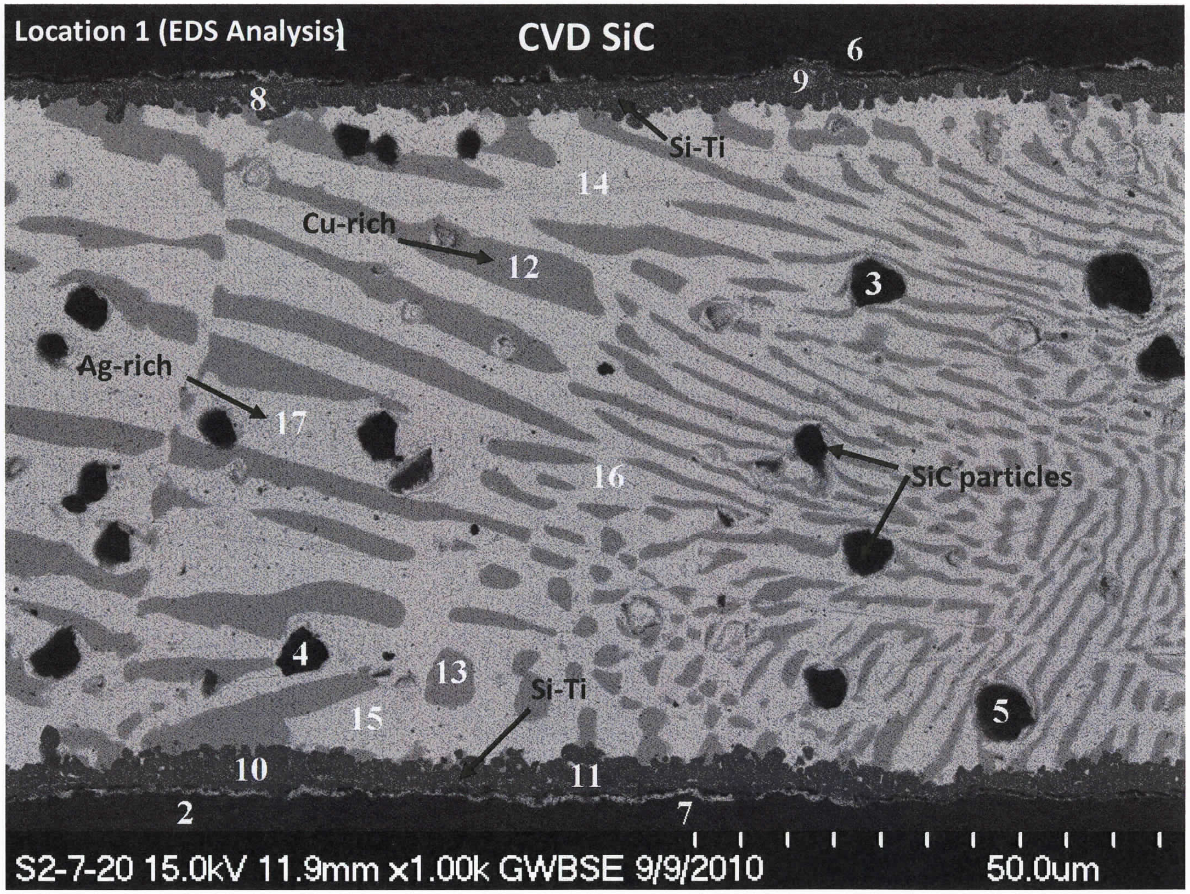


CVD SiC/Ticusil (5% SiCp)/CVD SiC

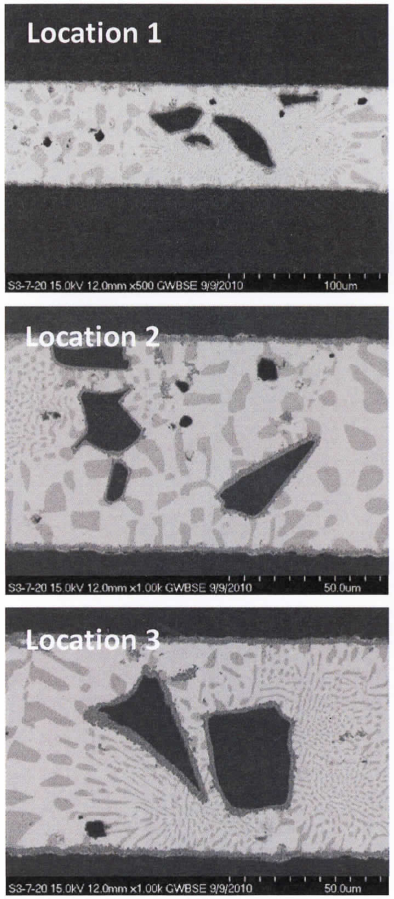


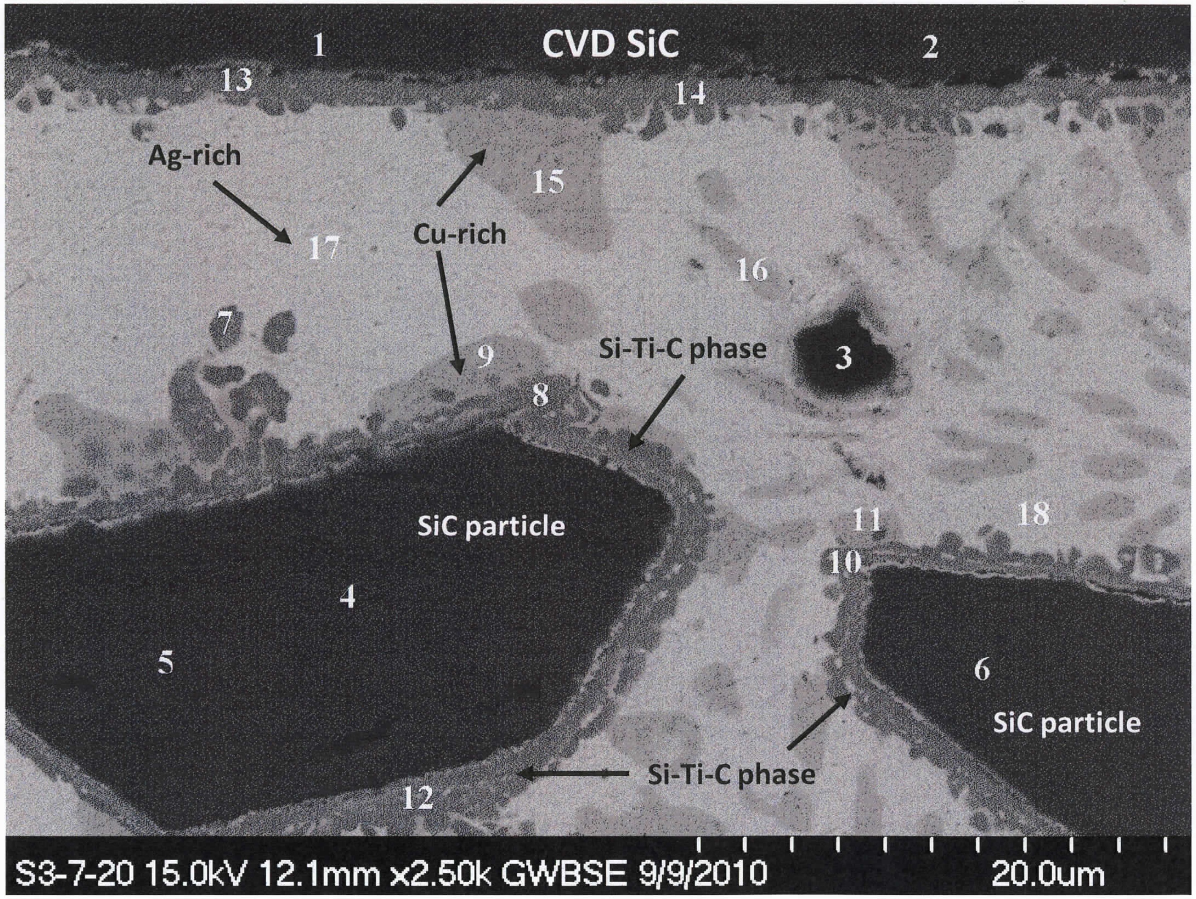




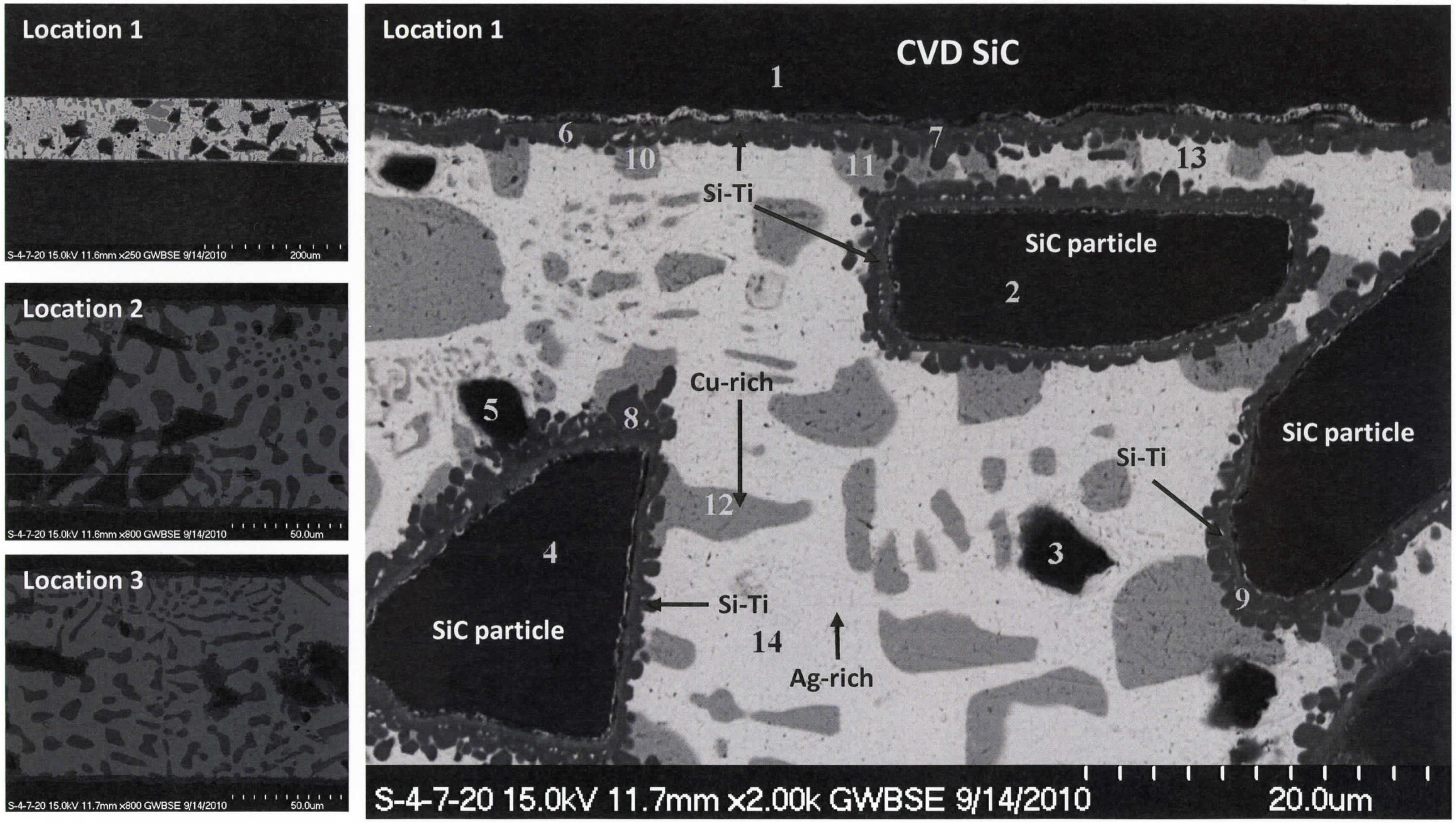


CVD SiC/Ticusil (10% SiCp)/CVD SiC



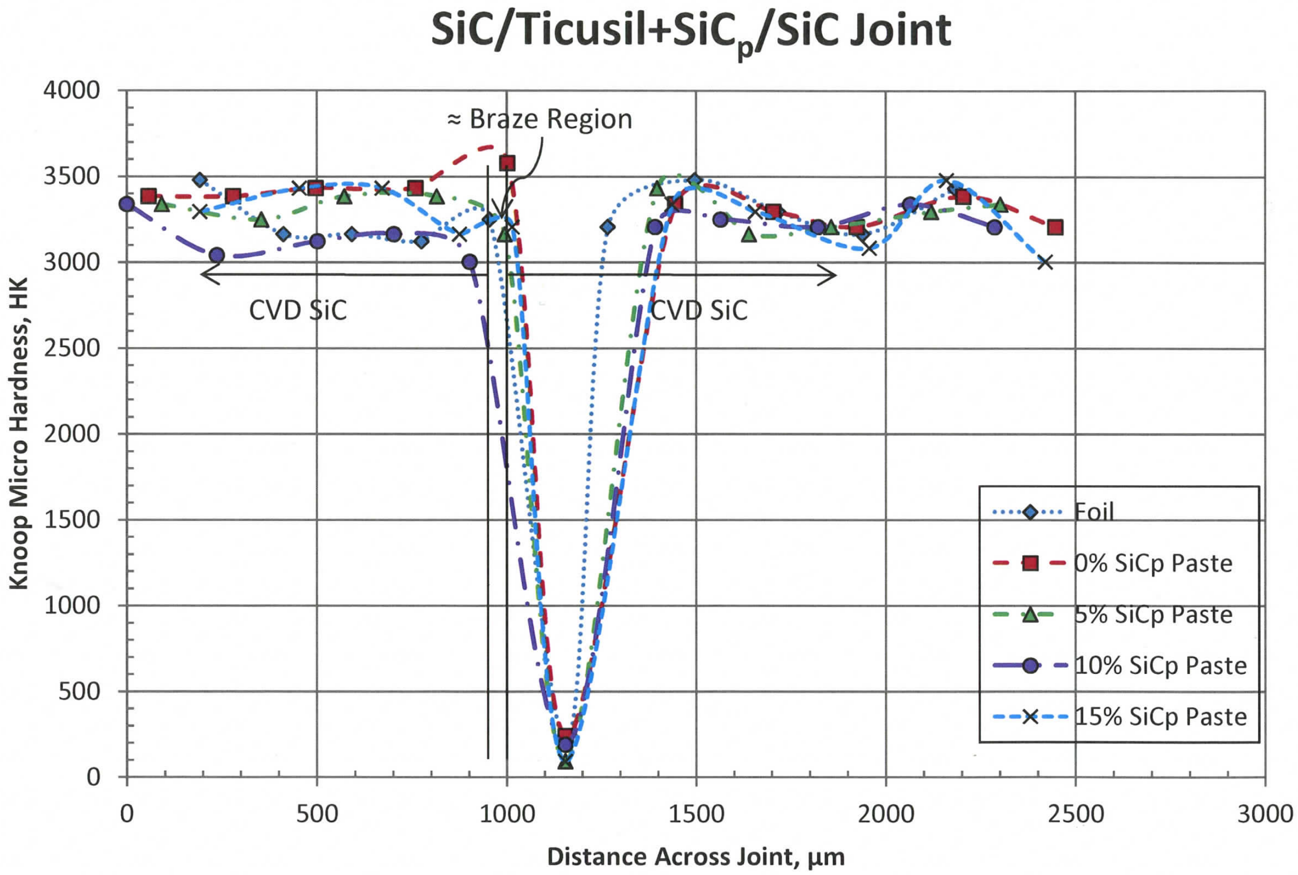


CVD SiC/Ticusil (15% SiCp)/CVD SiC



Effect of SiC Dispersion on Joint Hardness

(Ticusil Braze)



- ***The small braze thickness allowed only a single indentation to be made within the braze.***
- ***No measureable effect of SiC dispersion on joint hardness was noted***

Projected CTE Decrease with SiC Dispersion in Braze

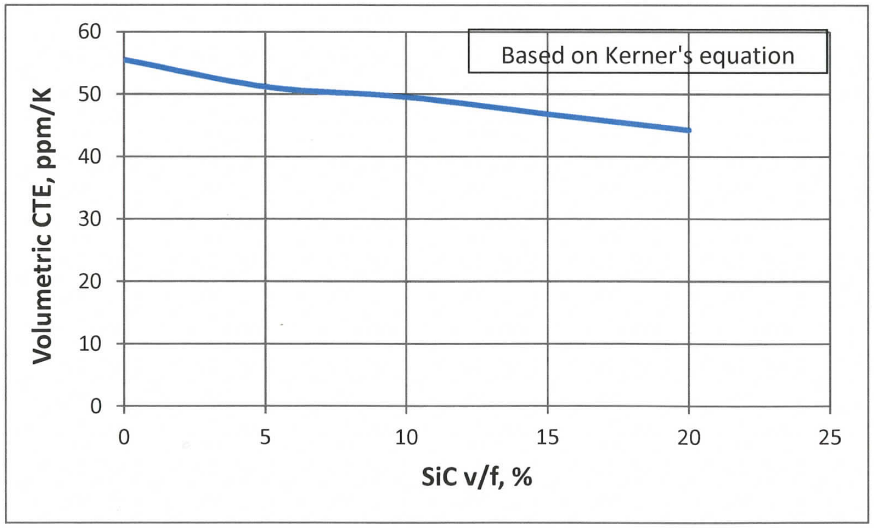
Kerner's equation for volumetric CTE of a matrix containing particles:

$$\alpha_C = \alpha_m V_m + \alpha_p V_p - (\alpha_m - \alpha_p) V_m V_p \left[\frac{(1/K_m) - (1/K_p)}{(V_m/K_p) + (V_p/K_m) + (3/4G_m)} \right]$$

(E.H. Kerner, Proc. Phys. Soc., B69, 1956, 808)

α_c = volumetric CTE of composite
α_m = volumetric CTE of matrix
α_p = volumetric CTE of particulate
V_m, Vp = volume fractions of matrix and particulate
K_m, Kp = Bulk moduli of matrix and particulate
G_m = Shear modulus of matrix

$\alpha_m = 55.5 \times 10^{-6}\ K^{-1}$
$\alpha_p = 12.3 \times 10^{-6}\ K^{-1}$
$K_m = E/[3(1-2\nu)] = 57GPa,\ E = 85\ GPa,\ \nu \sim 0.25$
$K_p = 203\ GPa$
$G_m \sim 0.4E = 34\ GPa$



At 15 v/f SiC in Ticusil braze, a 16% decrease is projected in the CTE of Ticusil

Reaction and Interface Formation

- Formation of TiC and titanium silicides ($TiSi_2$, Ti_5Si_3, $Ti_5Si_3C_x$) is thermodynamically feasible ($\Delta G<0$)
- Based on ΔG calculations, $TiSi_2$ and Ti_5Si_3 are less probable than TiC
- Titanium silicides could also form during solidification following SiC dissolution in braze and melt saturation with Si

Microstructural Observations

- SiC particles are randomly distributed in Ticusil and well-bonded
- Evidence of reaction of Ti with SiC and Ti-Si-C phase formation at interface
- SiC particles with Ti-rich interface aid nucleation of Cu-rich secondary phases
- Loss of Ti in reaction with SiC particulates did not impair bonding with SiC substrate
- Reinforcing AgCuTi braze with SiC can

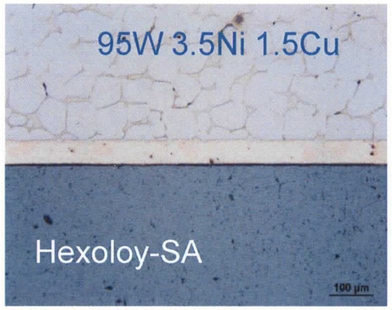
  - reduce the CTE mismatch and residual stresses
  - aid bonding
  - enhance braze strength

Observations

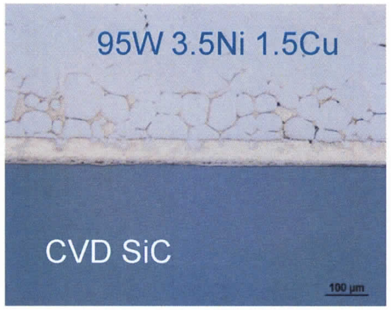
- Loss in braze ductility from SiC dispersion and ability to accommodate stress may be compensated by CTE decrease
- Excessive addition of SiC particles may induce pores in the joint
- Fine SiC causes clustering, metal-starved cavities and sites for crack initiation
- At very high SiC content, more Ti in filler may be used up in reaction with SiC particulates, leading to thinner reaction layer at substrate interface and affecting the joint strength

Bonding of Silicon Carbide-Metals (*ongoing…*)

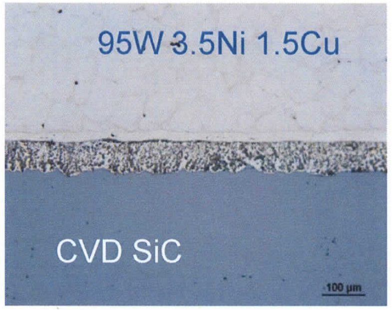
Cusil ABA



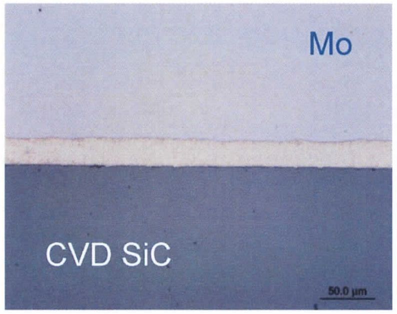
Ticusil



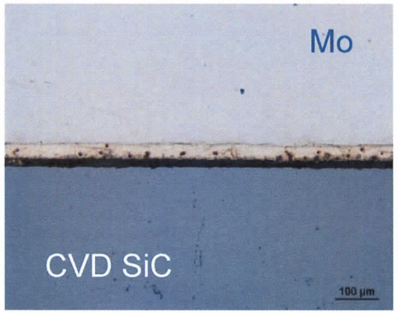
Copper ABA



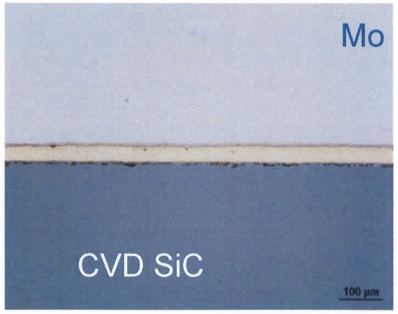
Cusil ABA



Ticusil



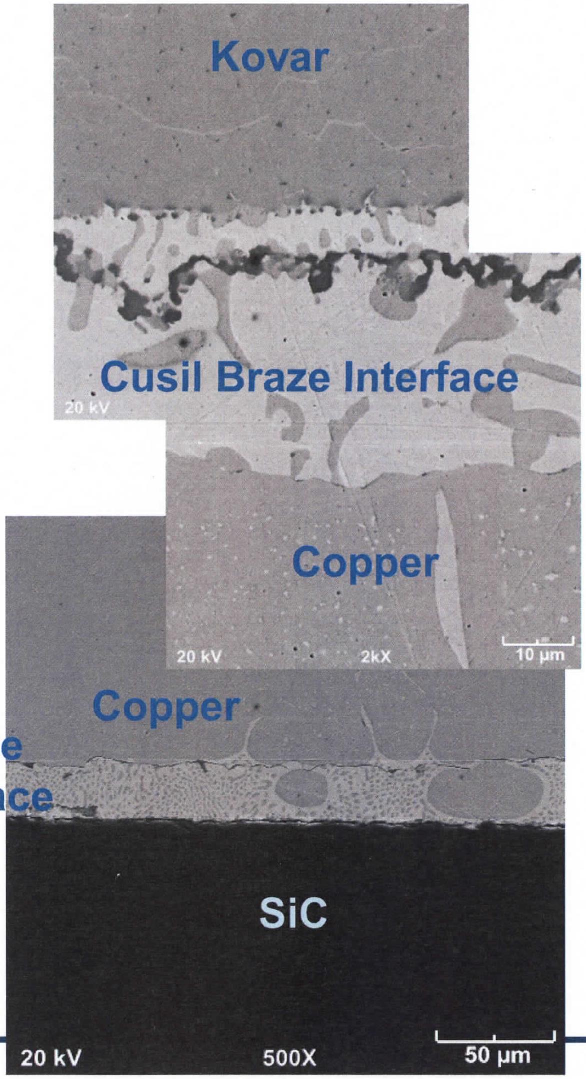
Copper ABA



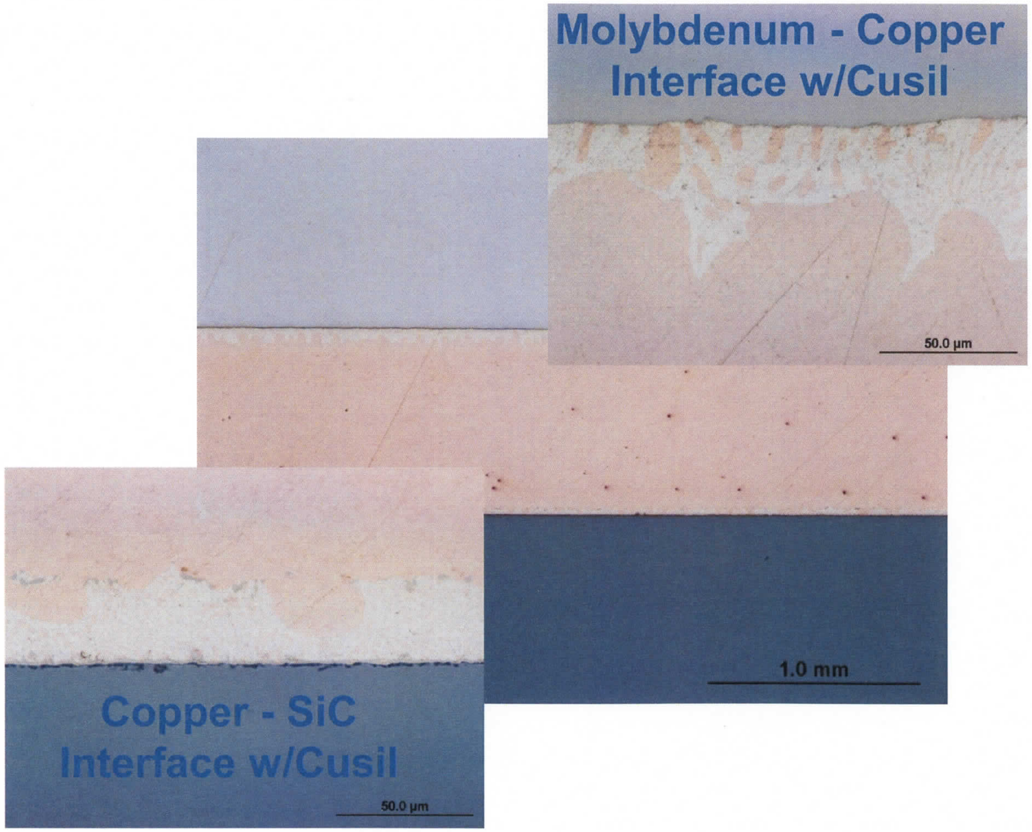
- **SiC brazed to SiC, Ti, W, Ni, and Mo using Ag-Cu-Ti and Cu-Si-Al-Ti brazes**
- **Multilayer joints created to manage CTE induced residual stresses**
- **SiC-to-metal joints created using diffusion bonding were also extensively characterized for fuel injector application**

Brazing of SiC to Kovar and to Molybdenum with Cusil ABA Foil/Copper/Cusil ABA Foil

Kovar / Cusil ABA / Copper / Cusil ABA / SiC

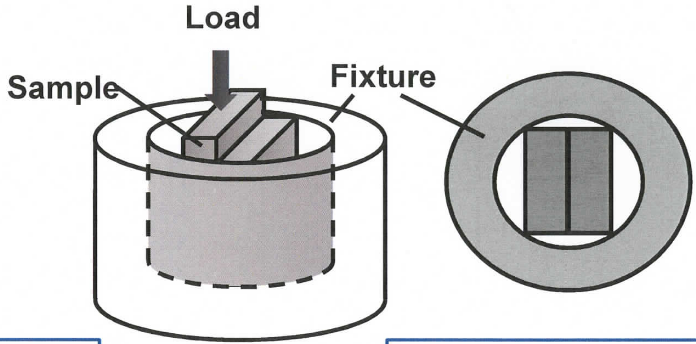


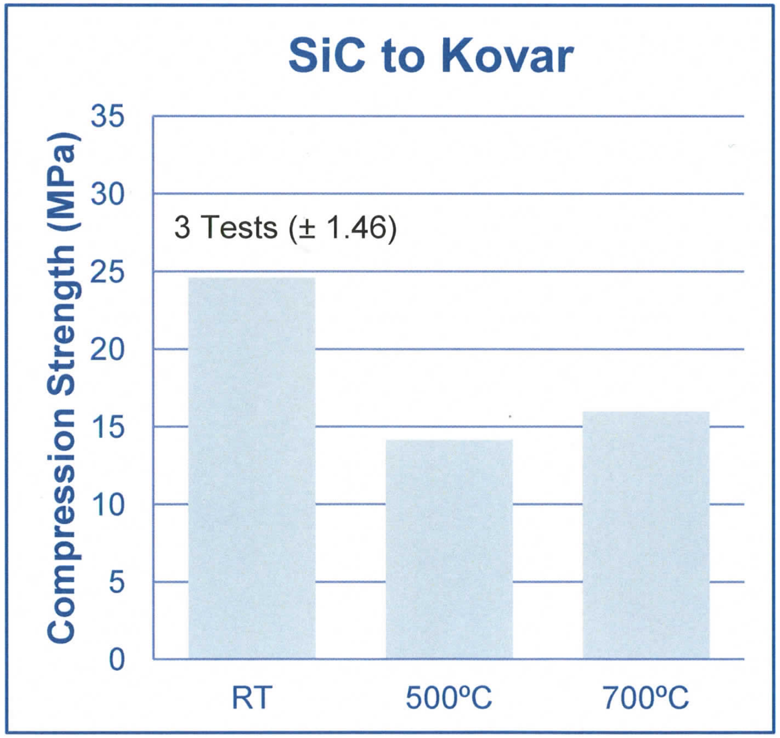
Molybdenum / Cusil ABA / Copper / Cusil ABA / SiC

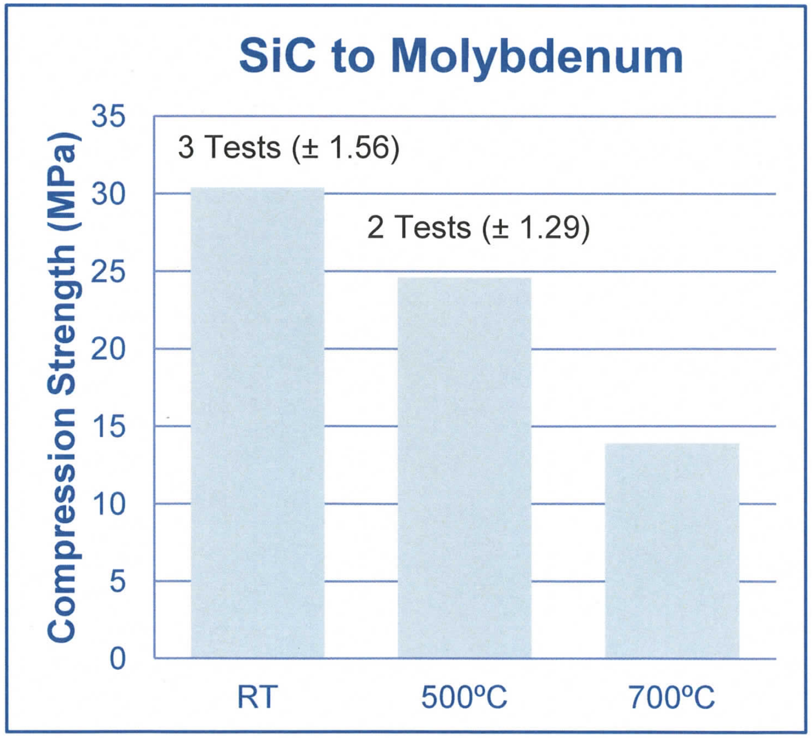


Shear Testing of SiC to Kovar and SiC to Molybdenum Joints with Copper and Cusil ABA Foil

- Testing Conditions
  - Crosshead speed: 0.5mm/min
  - Air
  - RT, 500°C, 700°C







Summary and Conclusions

- Ti containing active brazes offer excellent wetting of the SiC substrates.
- SiC particulate reinforcements in Ticusil braze alloys can be used to reduced the thermal expansion and enhance braze strength.
- Copper interlayers proved to be capable of absorbing residual stresses introduced from brazing and acted as a diffusion barrier preventing unwanted silicide and carbide formation.
- Initial mechanical testing shows that Mo-SiC joints brazed with a copper interlayer are capable of high strength – failure typically occurred within the ceramic.

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