FABRICATION AND CHARACTERIZATION OF BRAZED JOINTS FOR SiC-METALLIC SYSTEMS UTILIZING REFRACTORY METALS

ABSTRACT

Metal to ceramic joining plays a key role for the integration of ceramics into many nuclear, ground and aero based technologies. In order to facilitate these technologies, the active metal brazing of silicon carbide (CVD β-SiC, 1.1 mm thick, and hot-pressed α-SiC, 3 mm thick) to the refractory metals molybdenum and tungsten using active braze alloys was studied. The joint microstructure, composition, and microhardness were evaluated by optical microscopy (OM), scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), and Knoop hardness testing. The braze alloys, Cusil-ABA, Ticusil and Copper-ABA, all formed sound joints with excellent wetting and chemical bonding with the SiC substrate. Despite the close thermal expansion match between the metal substrates and SiC, hairline cracks formed in α-SiC while β-SiC showed no signs of residual stress cracking. The use of ductile interlayers to reduce the effect from residual stresses was investigated and joints formed with copper as an interlayer produced crack free systems utilizing both CVD and hot-pressed SiC.

Fabrication and Characterization of Brazed Joints for SiC-Metallic Systems Utilizing Refractory Metals

Bryan Coddington[1], Rajiv Asthana[1], Michael C. Halbig[2] and M. Singh[3]

1 – University of Wisconsin-Stout, Menomonie, WI
2 – NASA Glenn Research Center, Cleveland, Ohio
3 – Ohio Aerospace Institute, Cleveland, Ohio

STOUT
UNIVERSITY OF WISCONSIN

OAI
Ohio Aerospace Institute

35th International Conference and Exposition on Advanced Ceramics and Composites, Daytona Beach, Florida, January 23rd – 28th , 2011.

Outline

- **Application**
  - Nuclear, Electronic, Ground and Aero Based Technologies
- **Experimental Procedure / Joint Processing**
  - Tungsten to SiC Brazing
  - Molybdenum to SiC Brazing
  - Challenges Encountered
- **Characterization**
  - Optical Microscopy
  - Knoop Microhardness Testing
  - Scanning Electron Microscopy (SEM) coupled with Energy Dispersion Spectrometry (EDS)
- **Summary and Conclusions**

Joining Applications for SiC Based Components

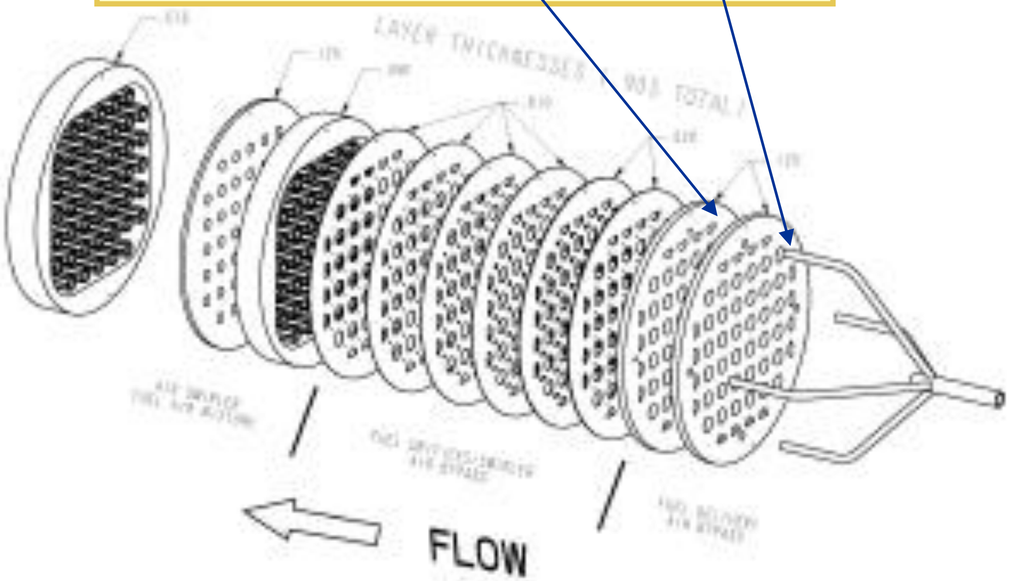
Applications for Mo & W Materials

- Electronic Components
  - Electrodes
  - Contacts
- Nuclear Applications
  - Radiation Shielding
  - Heat Shielding
- Aerospace Industries
  - Turbine Engine Components
    - Injectors
    - Turbines
  - Rocket Nozzles
- Ground Based Industries
  - Heating Elements
  - Medical Equipment

Fabrication and Testing of a MEMS Lean Direct Injector

Key Enabling Technologies:

- Brazing of SiC to Metallic Fuel Tubes
- Bonding of SiC to SiC



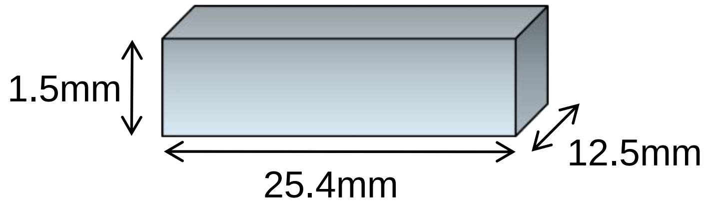
R. Tacina et al., TM-2002-211347

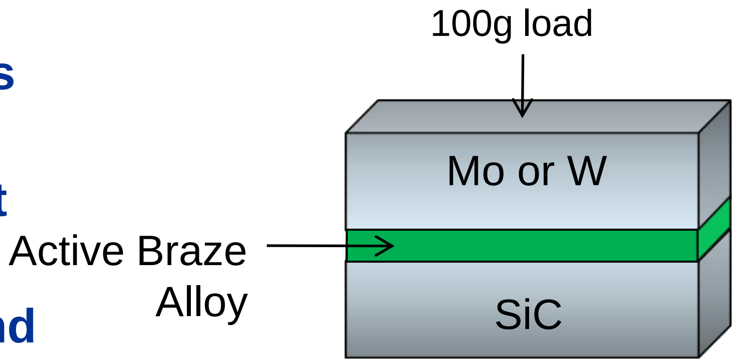
Develop and characterize the joining and integration technologies required for injector fabrication:

-**Brazing Metallic Tubes to SiC**

Key Enabling Technology – Metal/Ceramic Joining

- Experimental Procedure
  - **Substrates and braze foils were ultrasonically cleaned in Acetone for 10 minutes**
  - **Substrates were sandwiched together with a 100g load applied to the assembly**
  - **Heated to 10°C above the braze liquidus temperature**
  - **Isothermally held for 5 minutes under vacuum**
  - **Cooled to room temperature at 2°C/min**
  - **Mounted in epoxy, polished and the joints were characterized**





Key Enabling Technology – Metal/Ceramic Joining

Selected Substrate and Braze Properties

	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
Copper ABA	96	279	520	398	19.5	5.1	42
Cusil ABA	83	271	346	180	18.5	23	20
Ticusil	85	292	339	219	18.5	29	28
SiC	466	-	-	300	4.0*	-	-
Molybdenum	330	-	350	138	6.5*	-	-
Tungsten	310	590	758	33	4.4**	-	7
Nickel	190	150	345	67	13.5	-	47
Copper	117	70	220	391	16.9	-	42

* 0 - 1000°C

** 20 - 400°C

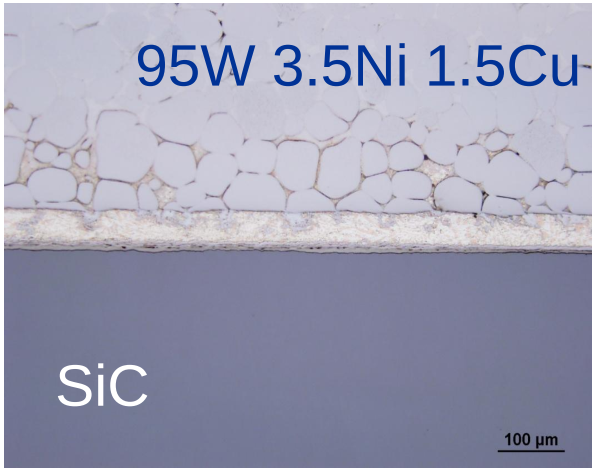
Key Enabling Technology – Metal/Ceramic Joining

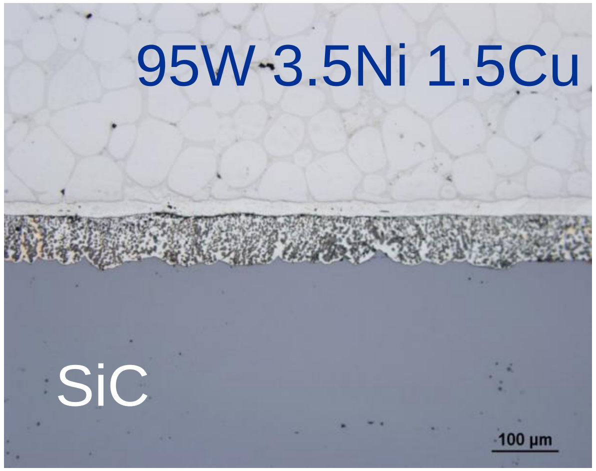
Challenges

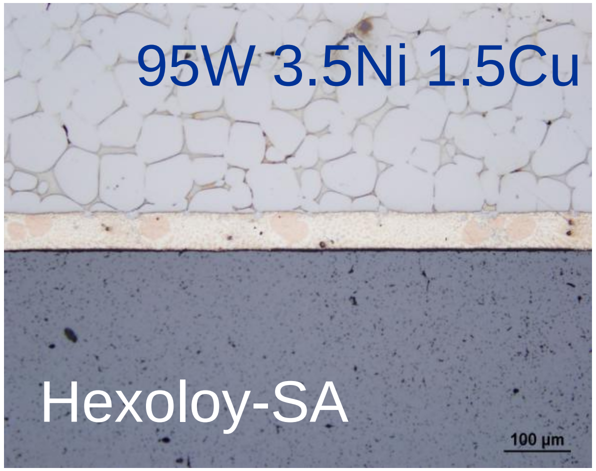
- **Wetting of the Ceramic Substrate**
  - **Active Braze Alloys – Ti**
    - Cusil ABA – 63Ag 35.25Cu 1.75Ti
    - Ticusil – 68.8Ag 26.7Cu 4.5Ti
    - Copper ABA – 92.75Cu 3.0Si 2.0Al 2.25Ti
- **Managing residual stresses due to differing Coefficients of Thermal Expansion (CTE)**
  - **Warping, Delamination, Cracking in SiC Substrate**
  - **Ductile Interlayer Approach**
- **Formation of Silicides**
  - **Nickel and Titanium**

Tungsten-SiC Micrographs

Ticusil	Copper ABA	Cusil ABA

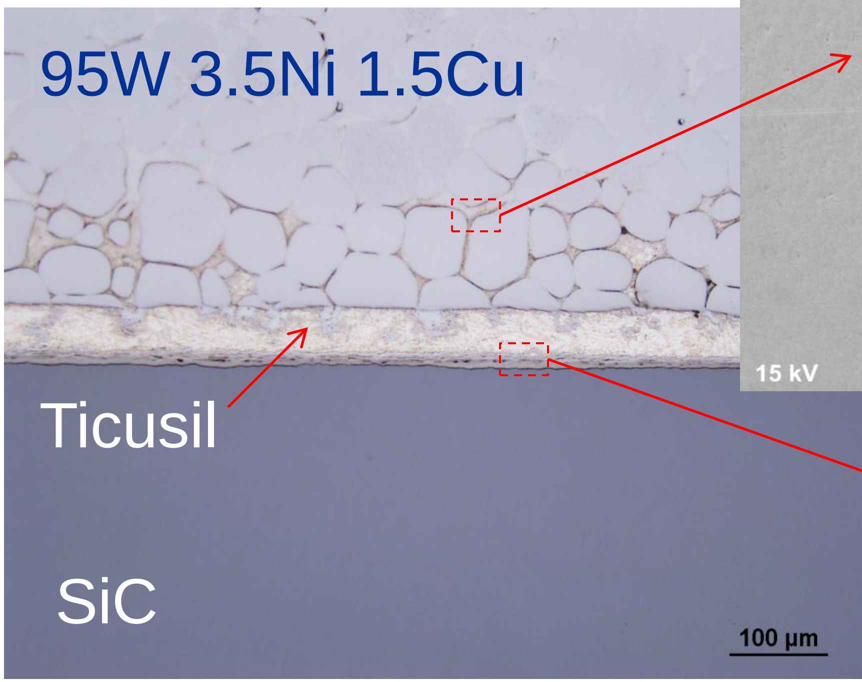


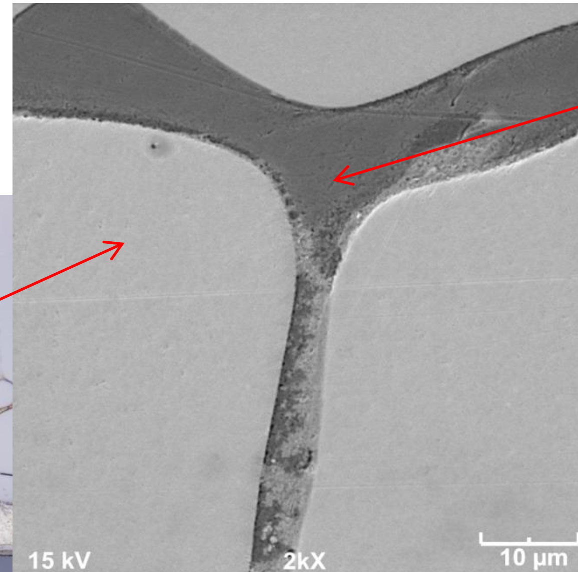




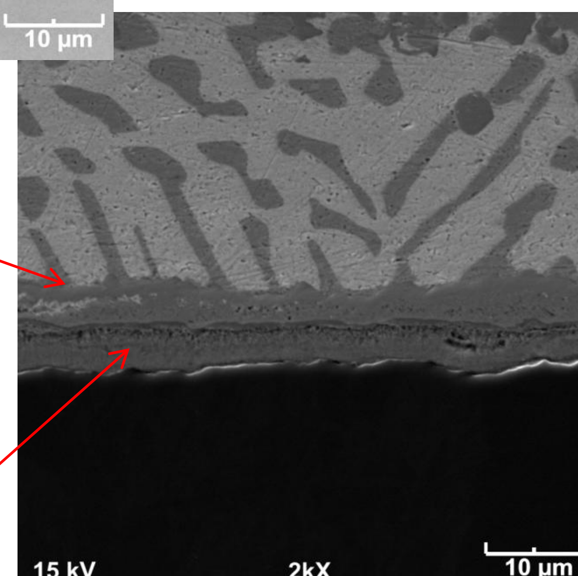
- All three brazes exhibited excellent wetting of the ceramic
- Nickel within the W alloy and the constituents of Ticusil interdiffused allowing nickel to react with the SiC substrate
- Samples brazed with Copper ABA also exhibited diffusion of the nickel within the W alloy allowing an interaction with SiC
- Samples formed with Hexoloy-SA showed nearly identical microstructure but with some minor stress cracking

EDS Analysis of Tungsten-SiC Joints



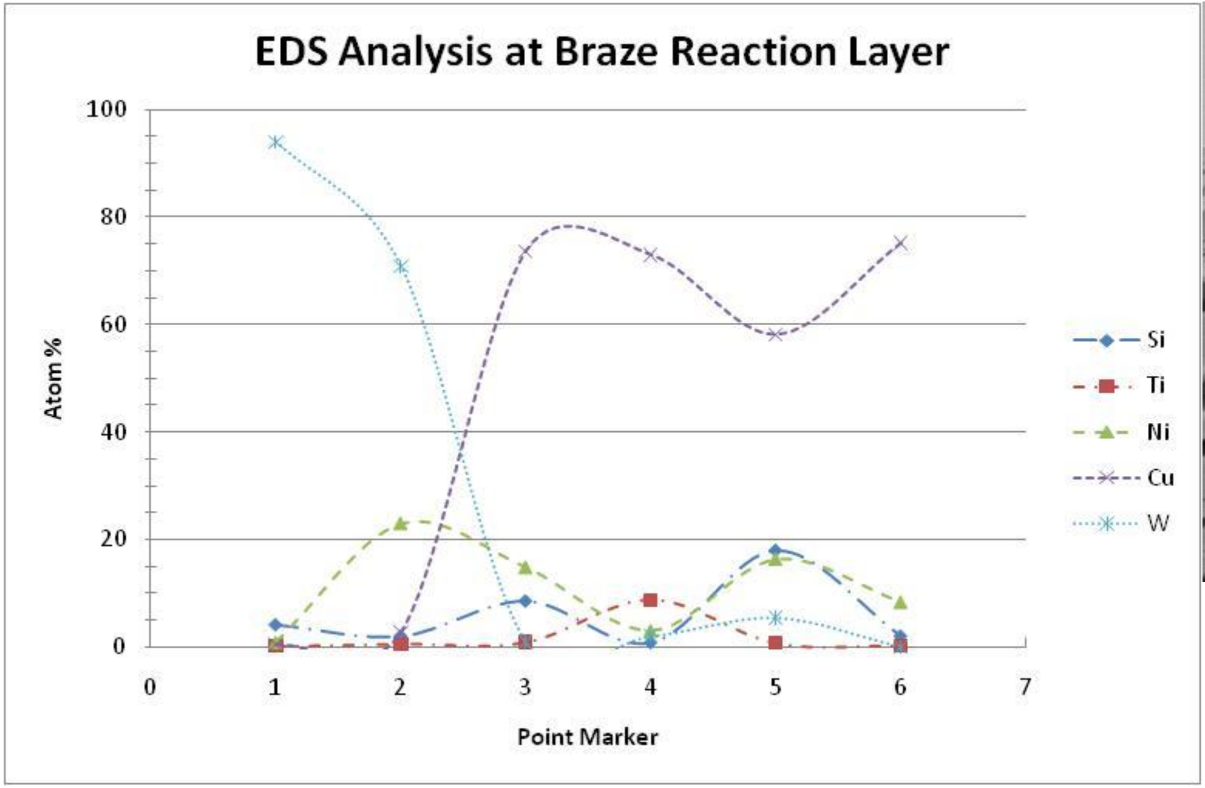


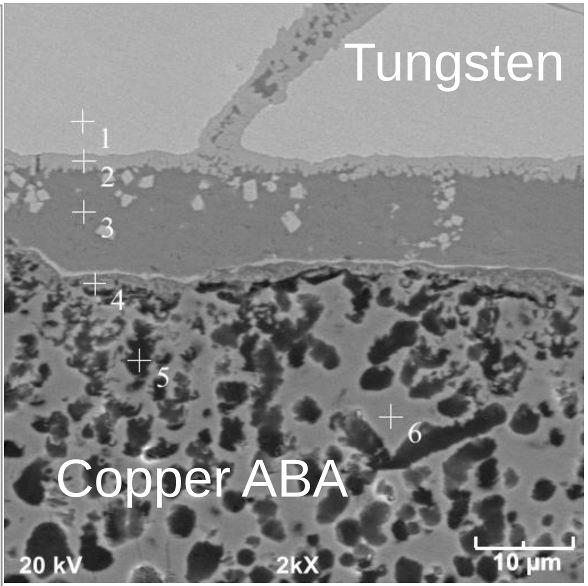
EDS analysis detected Ag along the grain boundaries of W



Nickel from the tungsten alloy diffused through the braze layer to form a possible nickel silicide at the SiC-braze interface

Energy Dispersive Spectroscopy (EDS) Analysis of Braze Reaction Layer at W - Copper ABA Interface

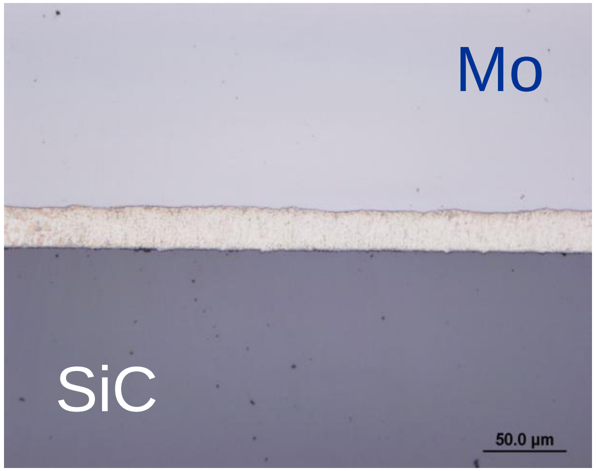




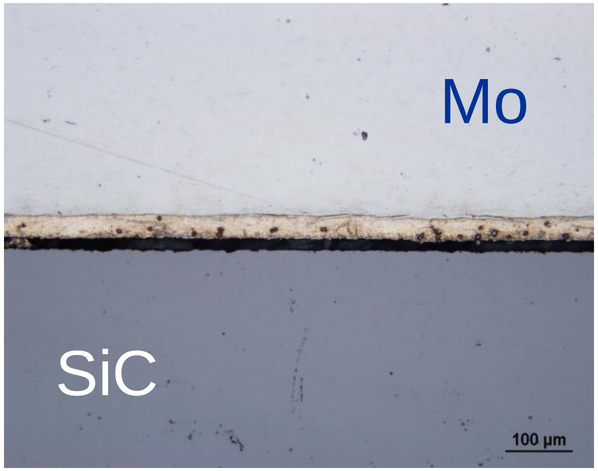
Point Marker	Si	Ti	Ni	Cu	W
1	4.181	0.135	0.797	0.492	93.991
2	2.07	0.574	22.92	2.825	70.832
3	8.599	0.881	14.752	73.708	0.902
4	0.87	8.761	2.966	72.938	2.015
5	18.009	0.695	16.286	58.147	5.424
6	2.075	0.261	8.279	75.169	0

Microstructures of Brazed Mo-SiC Joints

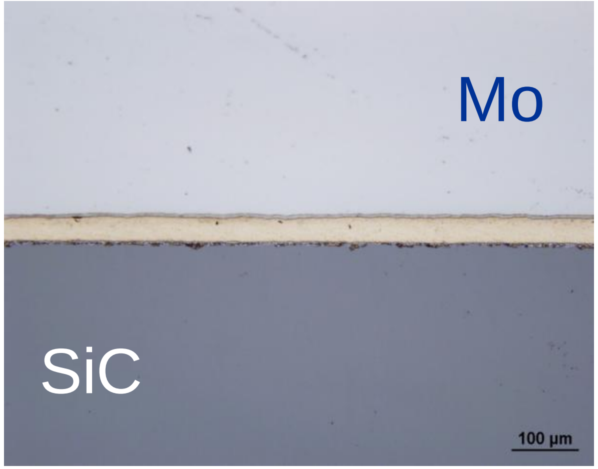
Cusil ABA



Ticusil



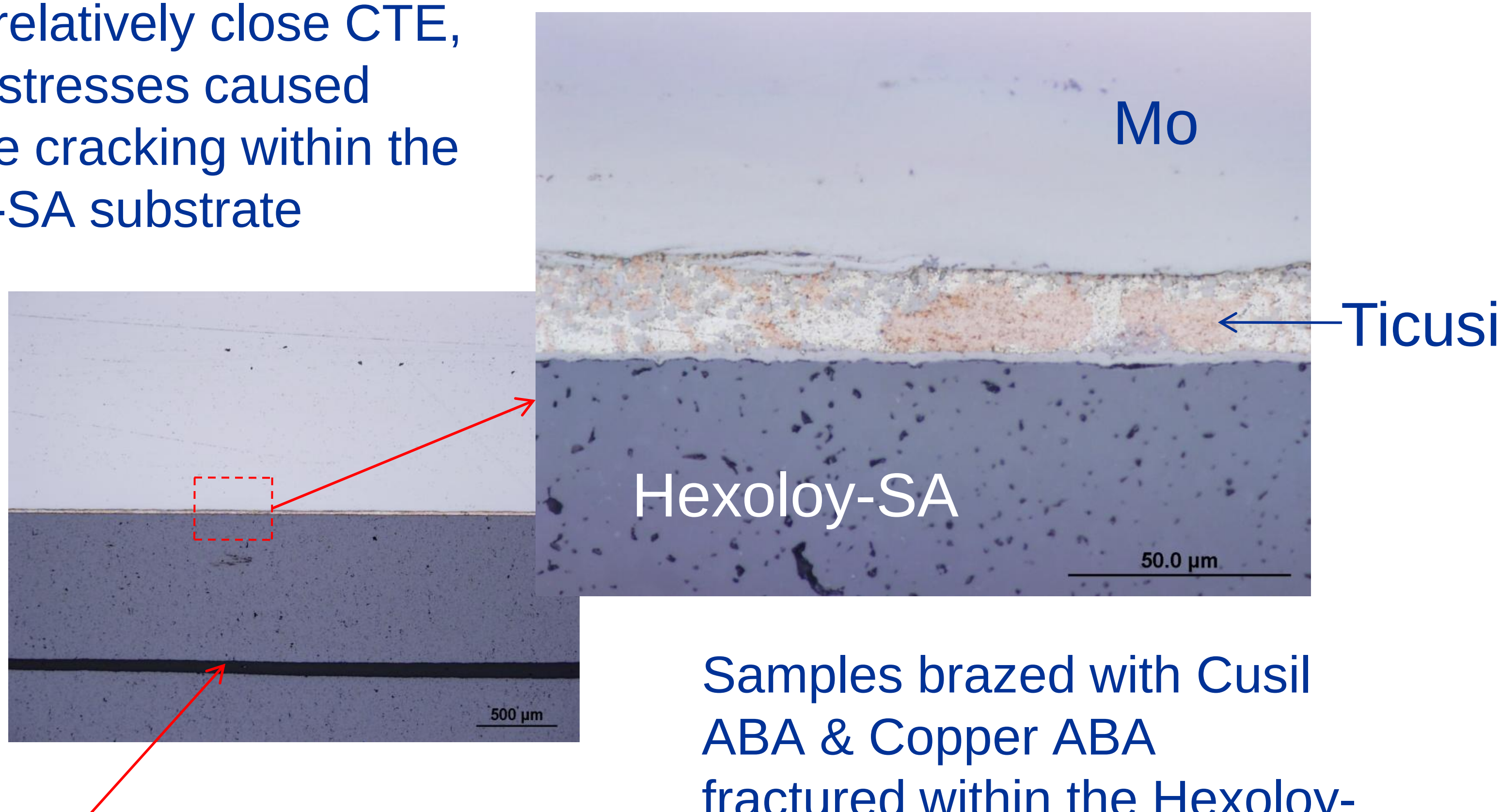
Copper ABA



- Cusil ABA appeared to wet both substrates very well
- Ticusil effectively wet the molybdenum substrate but a large gap was observed between the SiC substrate and the braze
- Copper ABA wet both the Molybdenum and the SiC very well.

Microstructures of Brazed Mo/Hexoloy-SA Joints

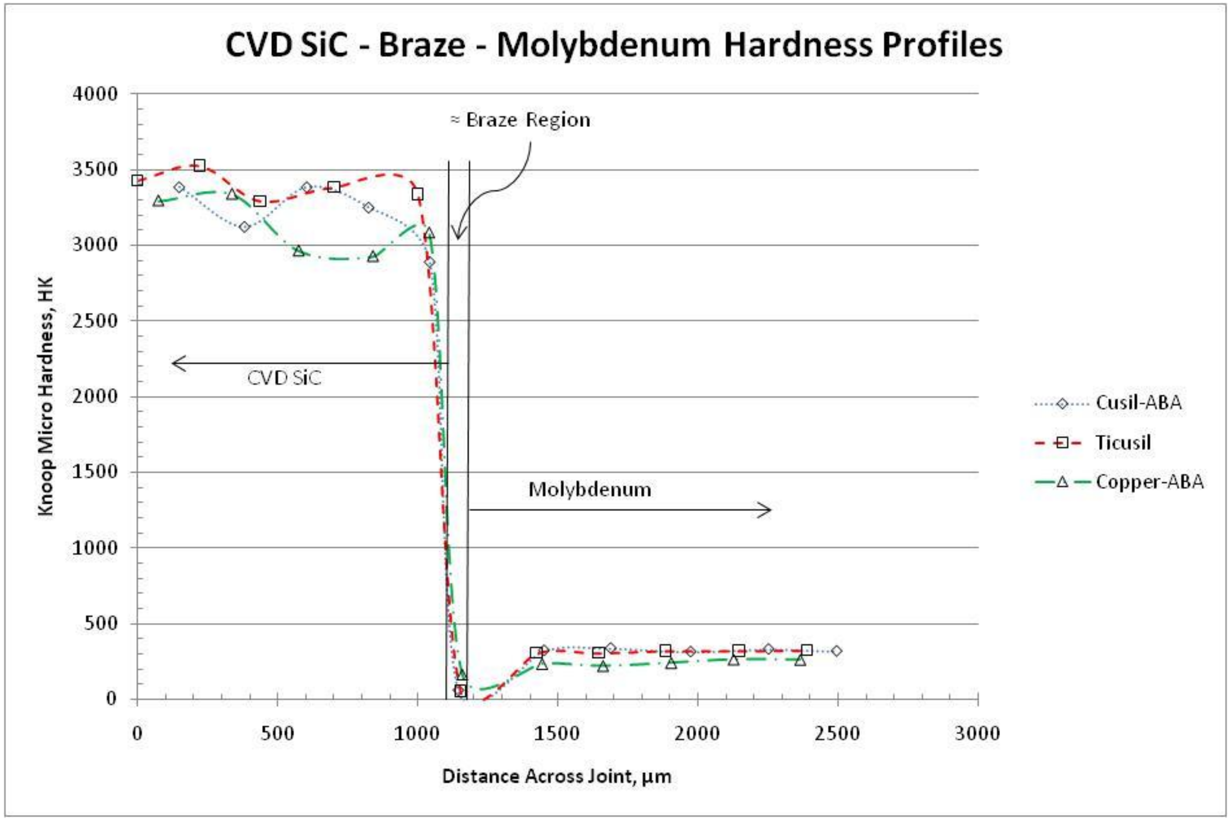
Despite relatively close CTE, residual stresses caused extensive cracking within the Hexoloy-SA substrate



Large crack in Hexoloy-SA substrate

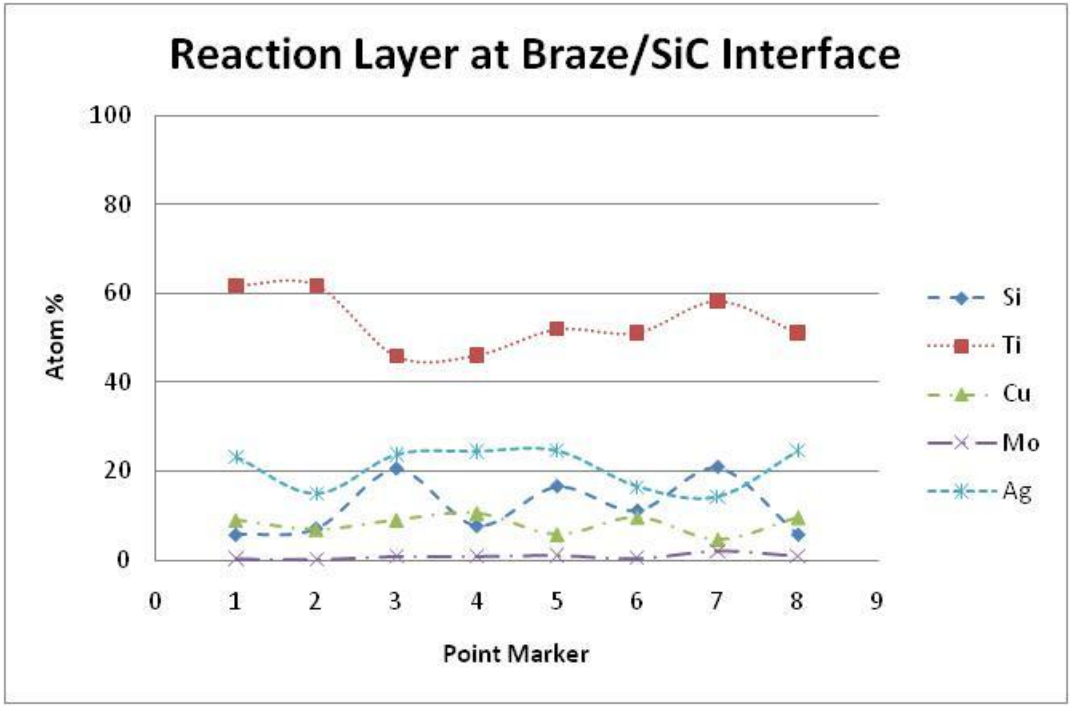
Samples brazed with Cusil ABA & Copper ABA fractured within the Hexoloy-SA substrate

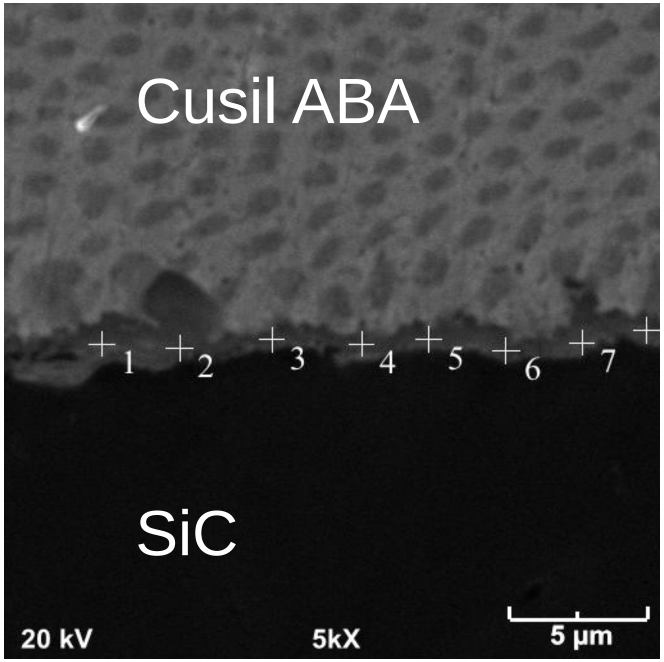
Knoop Microhardness Profiles for SiC-Mo Joints



	Cusil ABA		Ticusil		Copper ABA	
	µ	σ	µ	σ	µ	σ
CVD SiC	3205.8	185.5	3395	80.7	3121	167.8
Braze	58.8	0.4	55.35	0.25	180	16
Molybdenum	324.4	8.5	316.2	7.7	243.8	16.3

Energy Dispersive Spectroscopy (EDS) Analysis of SiC – Braze Interface





Point Marker	Si	Ti	Cu	Mo	Ag
1	5.619	61.78	8.971	0.41	23.22
2	6.993	61.794	6.907	0.26	14.837
3	20.438	45.856	9.003	0.907	23.797
4	7.615	46.105	10.49	0.899	24.497
5	16.509	51.92	5.81	1.115	24.647
6	11.091	51.165	9.567	0.448	16.485
7	20.826	58.256	4.712	2.056	14.149
8	5.684	51.096	9.52	0.963	24.578

Metallic Interlayer Approach Used to Absorb Residual Stresses

- SiC – Cusil ABA – Metal system exhibits good wetting and strong bonding
- Residual stresses are still a major concern
- Ductile interlayer approach is looked at to absorb thermal induced residual stresses
- Ceramic strain energy calculations are used to support ductile interlayer approach

Effect of Ductile Braze Layers on Residual Stresses

Ceramic Strain Energy Calculations

- Finite Element Method (FEM) used to calculate ceramic strain energy
- Analytical results accurate to within 1% of FEM – verified experimentally
- Eq. 1 can represent rectangular joints when interface area is approximated by a suitable r

1) $$U_{e,c} = \frac{\sigma_{YI}^2 r^3}{E_c}\{0.03\Pi_I + 0.11\varphi + 0.49\}$$

where 2) $$\Pi_I = \frac{(\alpha_m - \alpha_c)\Delta T E_I}{\sigma_{YI}}$$

and 3) $$\varphi = \left(\frac{\alpha_m - \alpha_I}{\alpha_c - \alpha_I}\right)^m$$

Calculated Values

Interlayer	Strain Energy mJ	Braze Temp. °C
Copper ABA	16.85	1044
Cusil ABA	15.69	825
Ticusil	18.25	910
Nickel	5.43	825
Copper	1.22	825

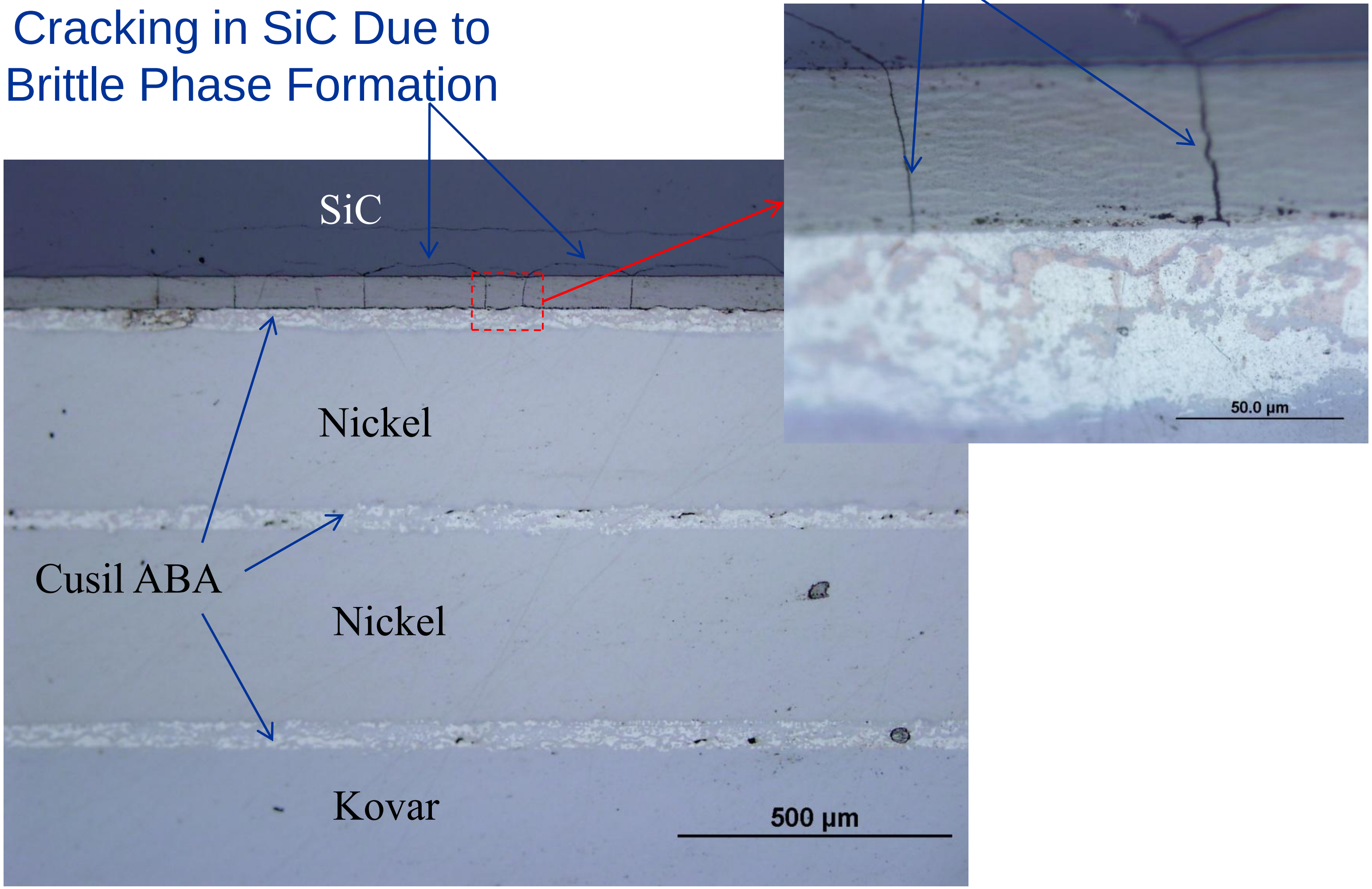
α_m = CTE for Molybdenum
α_c = CTE for SiC
α_I = CTE for interlayer
E_c = Elastic Modulus for SiC
E_I = Elastic Modulus for Interlayer
σ_{YI} = Yield Strength of Interlayer
ΔT = Braze Temp. – Room Temp.
r = radius of interface area

J.-W. Park, T.W. Eagar / Scripta Materialia 50 (2004) 555–559

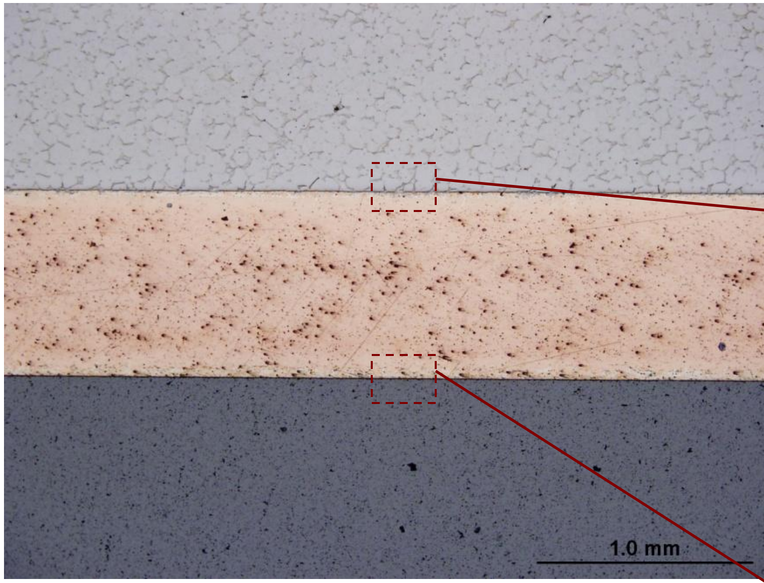
Formation of Brittle Phases Using Nickel Interlayers

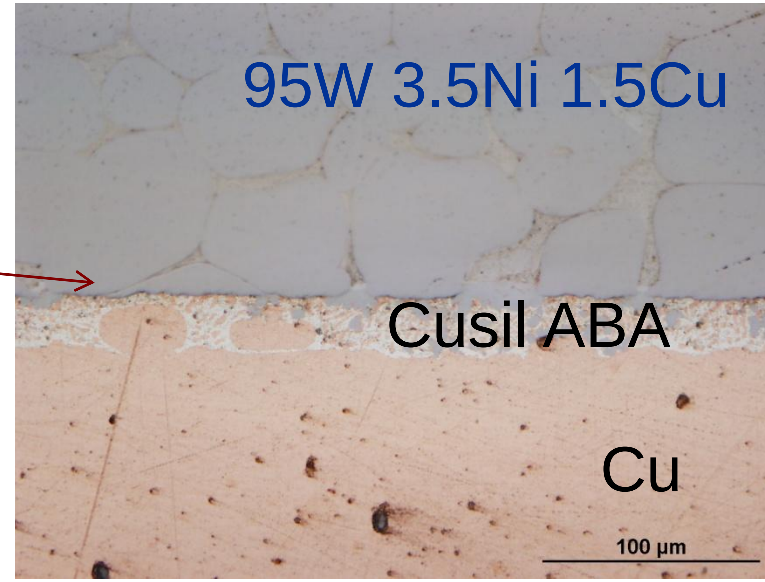
Cracking in Brittle Phase

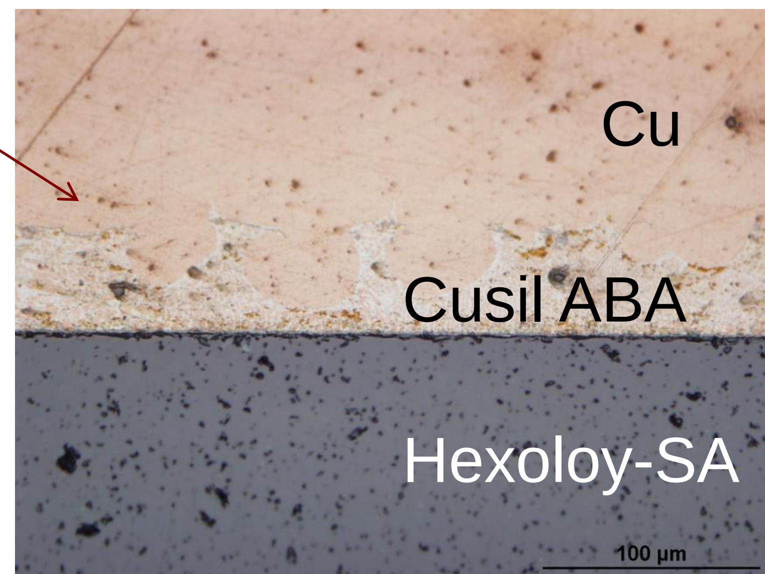
Cracking in SiC Due to Brittle Phase Formation



Microstructure of W-Cu-SiC Brazed Joint

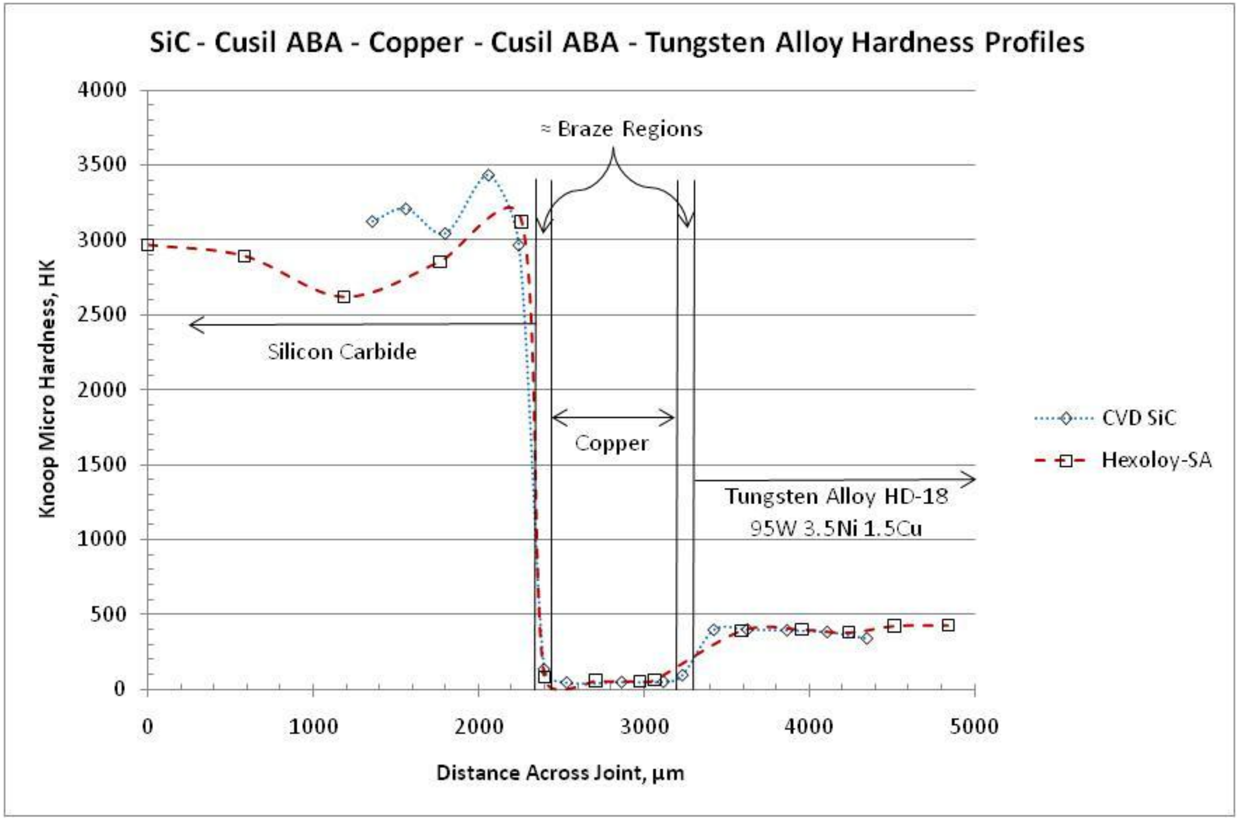






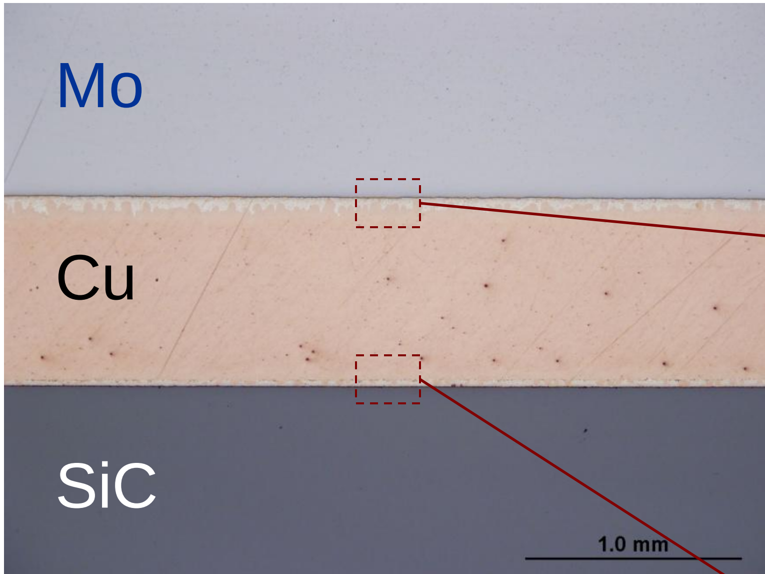
- Excellent wetting and bonding at all interfaces
- Copper interlayer has eliminated any residual stress cracking within the SiC substrate
- Copper acts as a possible diffusion barrier preventing unwanted silicide formation

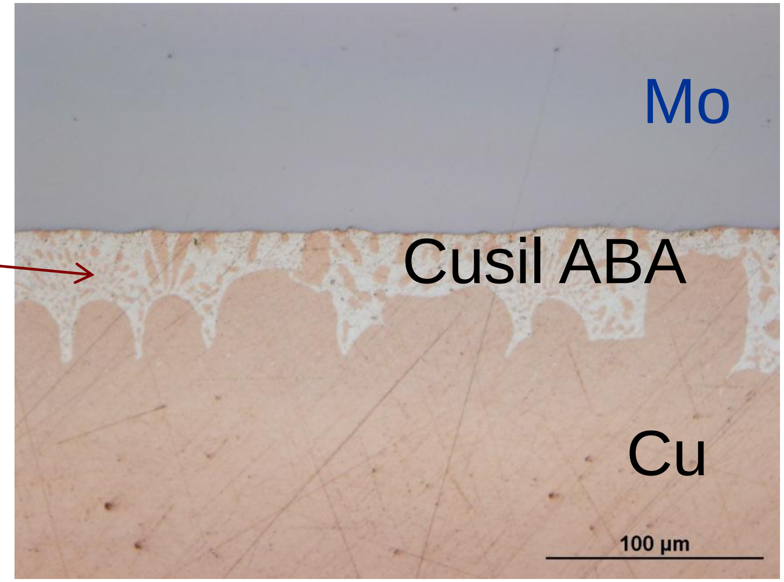
Knoop Microhardness Profiles for SiC-Cu-W Joints

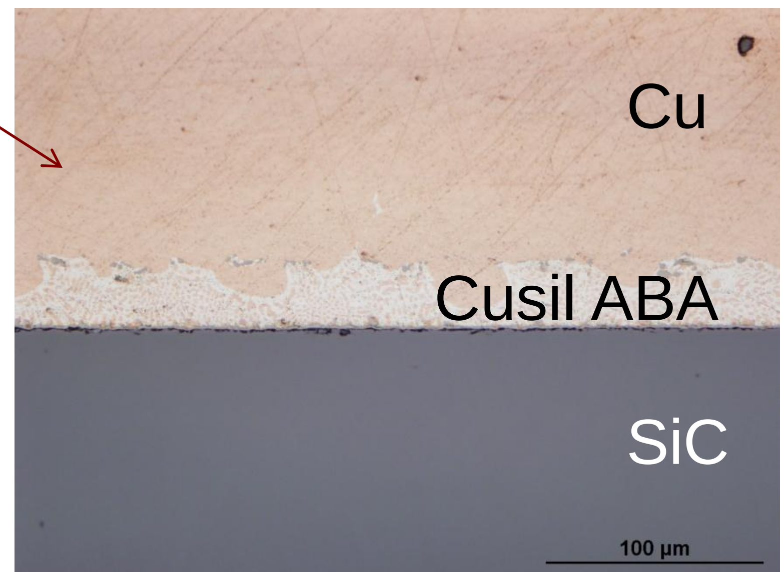


	CVD SiC		Hexoloy-SA	
	µ	σ	µ	σ
SiC	3153	160.7	2889.6	164.1
Cusil ABA	122	12.2	78.9	4.8
Copper	50.6	1.8	57.8	1.7
Cusil ABA	90.9	4.4	68.6	3.1
HD-18	384.6	22.0	405.8	24.0

Microstructure of Mo-Cu-SiC Brazed Joint

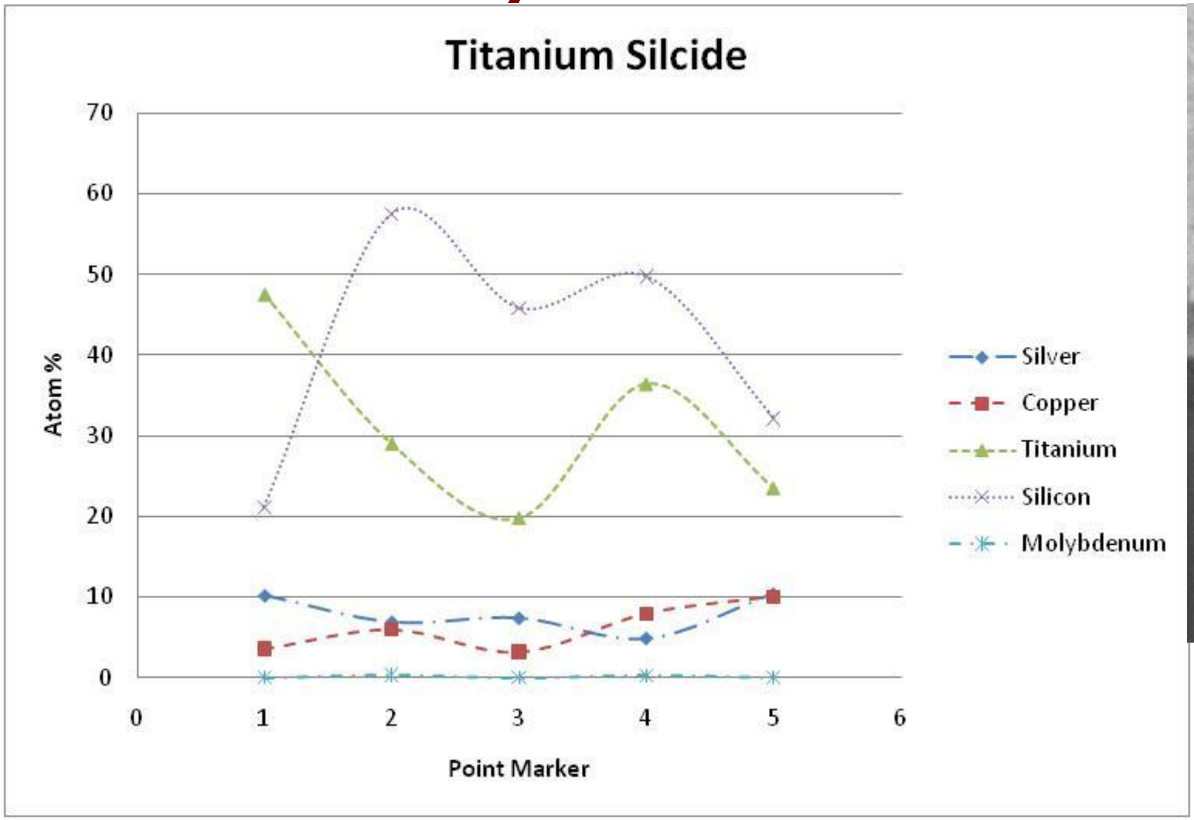


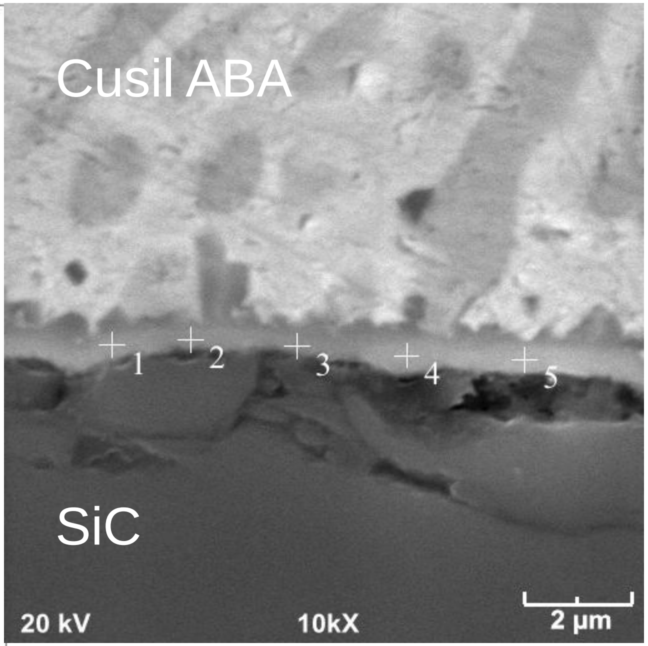




- Excellent wetting and bonding at all interfaces
- Copper interlayer has eliminated any residual stress crack within the SiC substrate
- Possible silicide formation at SiC/braze interface

Energy Dispersive Spectroscopy (EDS) Analysis of SiC – Braze Interface

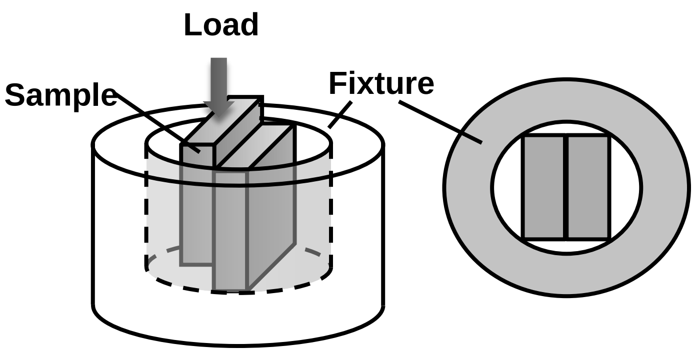


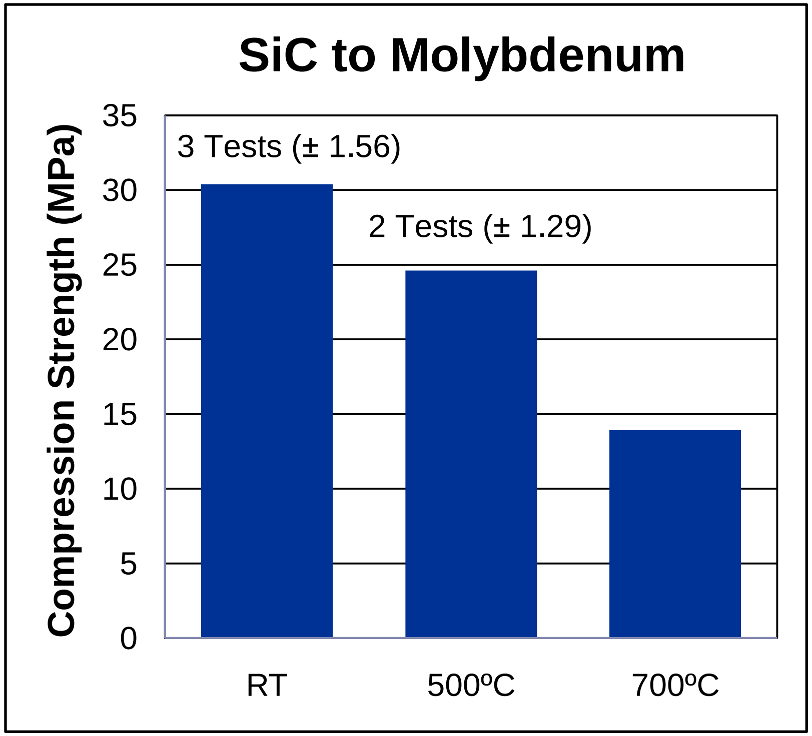


Point Marker	Ti	Cu	Mo	Ag	Si
1	47.621	3.583	0	10.232	21.138
2	29.07	5.955	0.312	6.949	57.545
3	19.831	3.22	0	7.441	45.855
4	36.483	7.931	0.299	4.928	49.773
5	23.535	10.015	0.018	10.442	32.151

Single Lap Offset (SLO) Shear Testing

- **Testing Conditions**
  - **Crosshead speed: 0.5mm/min**
  - **Air**
  - **RT, 500ºC, 700ºC**





The injector application requires a strength of about 3.45-6.89 MPa (0.5 - 1.0 ksi).

Summary and Conclusions

- Cusil ABA offered excellent wetting of the SiC and Metal substrates
- Brazing SiC to Molybdenum or Tungsten introduced residual stresses and caused cracking in the α-SiC substrate despite relatively small CTE mismatch.
- The use of nickel interlayers in a SiC system produced extensive diffusion of the nickel forming a possible nickel silicide
- 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

Acknowledgements

- One of the authors (Bryan C.) would like to thank LERCIP Program at NASA Glenn Research Center for summer research support and the University of Wisconsin-Stout for providing the necessary funding to present this research