Daytona Beach Conference, Abstract, American Ceramic Society

Bonding and Integration of C-C Composite to Cu-Clad-Molybdenum for Thermal Management Applications

R. Asthana[1 *], M. Singh[2],and T.P. Shpargel[3]

[1]Department of Engineering and Technology
University of Wisconsin-Stout
Menomonie, WI 54751, USA

[2]Ohio Aerospace Institute
NASA Glenn Research Center
Cleveland, OH 44135, USA

[3]ASRC Aerospace
Cleveland, OH 44135, USA

Abstract

Two- and three-dimensional carbon-carbon composites with either resin-derived matrix or CVI matrix were joined to Cu-clad-Mo using active Ag-Cu braze alloys for thermal management applications. The joint microstructure and composition were examined using Field-Emission Scanning Electron Microscopy and Energy-Dispersive Spectroscopy, and the joint hardness was characterized using the Knoop microhardness testing. Observations on the infiltration of the composite with molten braze, dissolution of metal substrate, and solute segregation at the C-C surface have been discussed. The thermal response of the integrated assembly is also briefly discussed.

---

* Currently with ASRC Aerospace, Cleveland, OH

Bonding and Integration of C-C Composites to Copper-Clad-Molybdenum for Thermal Management Applications

R. Asthana*, M. Singh, and T.P. Shpargel*****

*** University of Wisconsin-Stout**
Menomonie, WI 54751

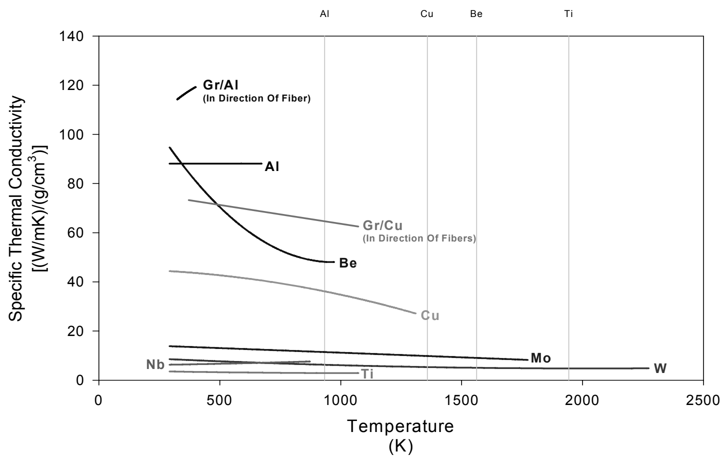
****Ohio Aerospace Institute**
NASA Glenn Research Center
Cleveland, OH 44135

***** ASRC Aerospace**
NASA Glenn Research Center
Cleveland, OH 44135

Overview

- Introduction and Background
- Experimental Procedures
  - Materials and Brazing
  - Characterization *(Microstructure, Microhardness)*
- Results and Discussion
  - Microstructure and Composition of Joints
  - Microhardness
  - Residual Stress and Thermal Considerations
- Concluding Remarks
- Acknowledgments

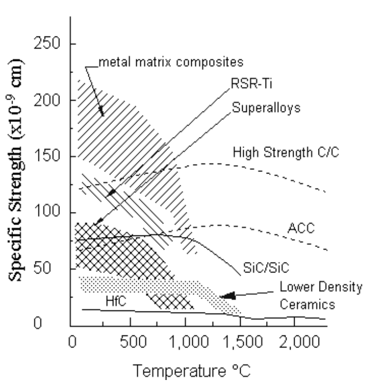
Thermal Properties of Some Typical Thermal Management Materials

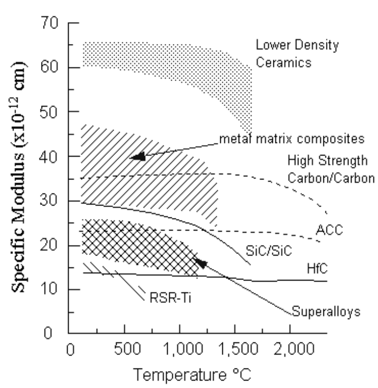


Operating Temperature Can Limit Material Choices

Thermal Management Material Properties

Carbon-Carbon Composites Provide Tremendous Advantage and Excellent Benefits for Thermal Management

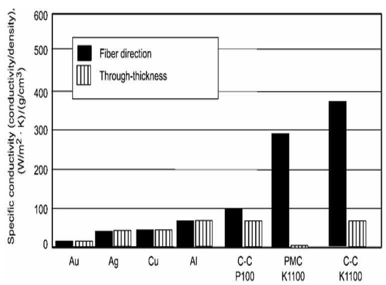




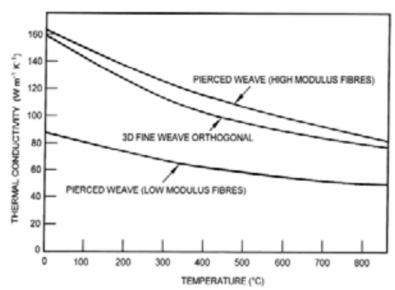
From: *"High thermal conductivity composites for passive thermal management," Metal Matrix Composites Information Analysis Center -Current Highlights, 8, 2 (1988).*

Application of Carbon-Carbon Composite Materials in Thermal Management Applications

Thermal conductivity of C/C composites strongly depends on the fiber type, architecture, and composite processing technology



Source: K. Kearns, Composites, ASM Handbook, Vol. 21 (2002) 1067-1070.

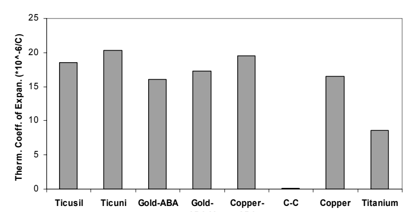


Source: Comprehensive Composite Materials, Vol. 4, 3 (2000).

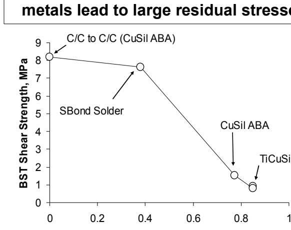
• *High modulus, high conductivity pitch based carbon fibers can be used to improve the thermal properties of C-C composites.*

Active Metal Brazing of Titanium to C/C Composites for Heat Rejection Systems

- Recently, we had joined C-C composite to Ti tubes for lightweight heat exchanger applications.
- Both direct bonding using braze layers and indirect bonding using a porous carbon foam (saddle material) and braze layers were employed.
- Excellent bonding of active braze to foam, C-C Composite, and Ti Tube occurred.
- Failure always occurred in Poco HTC (Saddle Material) indicating that bond strength exceeded the fracture strength of foam.



Large difference in CTE of C-C and metals lead to large residual stresses



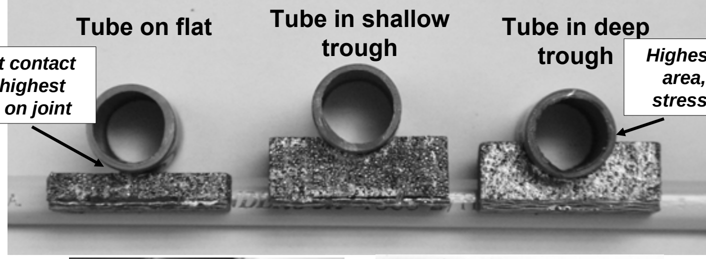
$\Delta\alpha = \alpha$ (Ti) – α (C/C)
ΔT = T (liquidus ~ processing) – 25°C

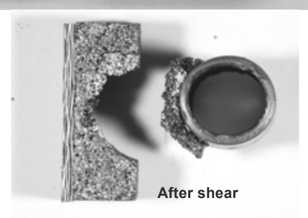
1. *M. Singh et al, Mater. Sci. Eng. A (in press).*
2. *M. Singh et al, Mater. Sci. Eng., A 412, 2005, 123-128.*
3. *G.N. Morscher et al, Mater. Sci. Eng., A 418(1-2), 2006, pp 19-24.*

Testing of Brazed Joints in Woven K1100 and P120 C/C Facesheet/Foam/Ti Tube

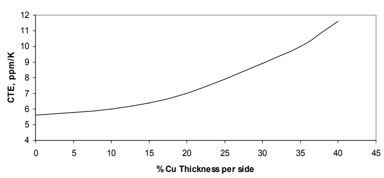
M. Singh et al, Mater. Sci. Eng., A 2008 (in the press)

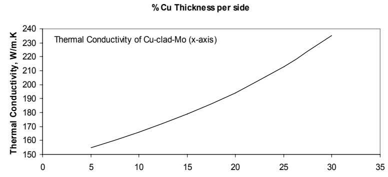
Specimens with different tube-foam contact areas were fabricated in order to vary braze contact area and stress applied to joints

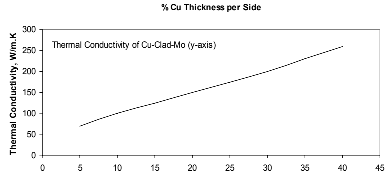




Why Copper Clad Molybdenum was chosen as a Thermal Management Material ?







(from Electronic Materials and Processes Handbook, C.A. Harper, McGraw-Hill, 2003)

- **Copper has excellent thermal conductivity (K for OFHC Cu: 401 W/m.K).**
- **CTE of Cu is high (16.5 ppm/K). Difficulty in joining to ceramic substrates.**
- **Low annealing temperature of Cu causes softening at moderate heat input.**
- **Cladding Mo with Cu lowers CTE and promotes thermoelastic compatibility with ceramics.**
- **Some loss of thermal conductivity (Mo: 138 W/m.K, Cu: 401 W/m.K).**
- **Small weight penalty (density of Cu: 8,900 kg.m^{-3}, density of Mo: 10,280 kg.m^{-3}).**

Objective

- **Develop brazing approaches for 2D and 3D C-C composites with resin and CVI matrices to Cu-clad-Mo using active braze alloys.**
- **Characterize the joint microstructure, composition, and microhardness behavior.**

Experimental Procedure: Materials and Properties

- **2-D and 3-D C-C composites (Resin +CVI Carbon matrix) –** ***Goodrich Corp., CA.***
- **C-C composites (Resin derived matrix) –** ***C-CAT, Inc., TX.***
- **Cu-clad-Mo plates (Cu-Mo-Cu ratio: 13%-74%-13%) –** ***H.C. Starck, Inc., MA.***
- **Active braze alloy (ABA) powders –** ***Morgan Advanced Ceramics, CA.***

Composition and Properties of Brazes

Braze (composition, %)	T_L, °C	T_S, °C	E, GPa	YS, MPa	UTS, MPa	CTE, $\times10^{-6}$ C^{-1}	% El.	K, W/m.K
Cusil-ABA® (63Ag-35.3Cu-1.75Ti)	815	780	83	271	346	18.5	42	180
Ticusil® (68.8Ag-26.7Cu-4.5Ti)	900	780	85	292	339	18.5	28	219

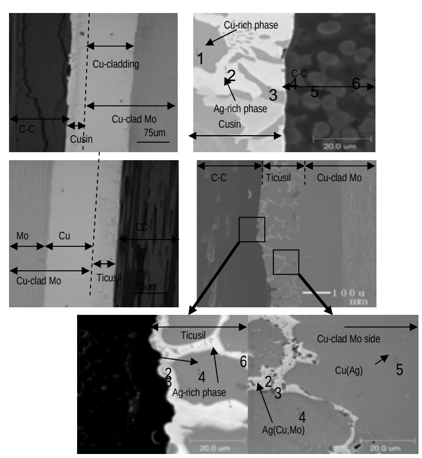
E: Young's modulus, YS: yield strength, UTS: tensile strength, CTE: coefficient of thermal expansion, %El: percent elongation, K: thermal conductivity

Experimental Procedure

- **Substrates cut into 2.54 cm x 1.25 cm x 0.25 cm plates and ultrasonically cleaned in acetone for 15 min.**
- **Braze powders mixed with glycerin to dough-like consistency and braze paste manually applied to C-C surface.**
- **Assembly heated under vacuum (~10^{-6} torr) to 15-20°C above braze T_L. After 5 min. soak, slowly cooled (~5ºC per min.).**
- **Brazed joints mounted in epoxy, ground, polished, and examined using optical microscopy and Field Emission Scanning Electron Microscopy (Hitachi 4700) coupled with EDS.**
- **Microhardness (Knoop indenter) on Struers Duramin-A300 machine (200 g load, 10 s). Four-to-six scans across each joint.**

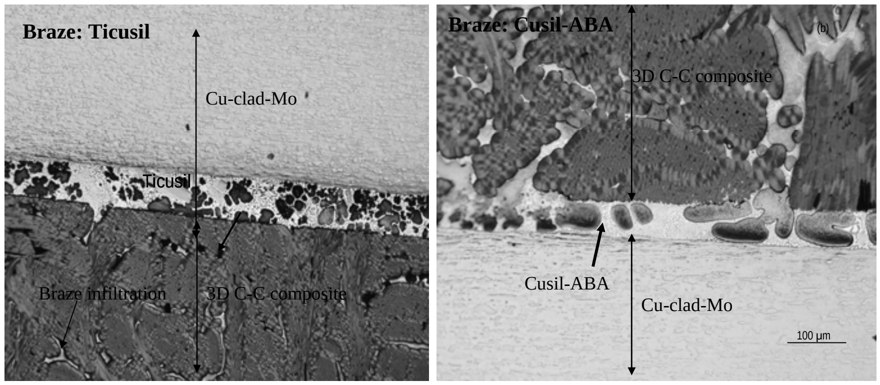
C-C Composite/Cu-Clad-Mo Joints for Thermal Management Applications

- **Resin-derived C-C composites brazed to Cu-clad Mo using active braze alloys (ABA).**
- **Good metallurgical bonding at joints, with some dissolution, diffusion, and solute redistribution.**
- **Ti segregated at C-C surface. Cu-ABA joints displayed the largest Ti concentrations at joint.**
- **Microhardness gradients exist at joints.**
- **C-C/Cu-clad Mo systems may have potential for thermal management applications.**



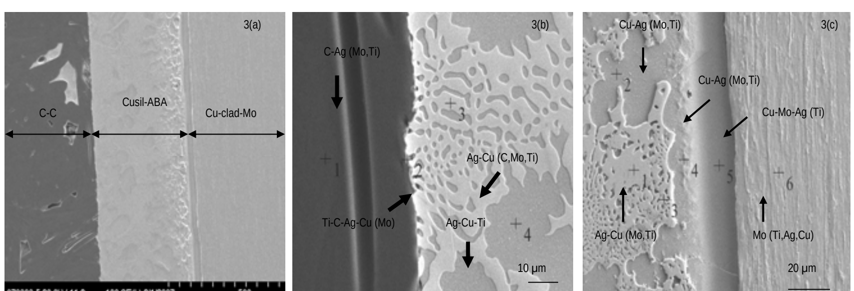
M. Singh, R. Asthana, T. Shpargel, Mater. Sci. Eng., A 452-453, 2007, 699-704

3-D C-C Composite/Cu-clad-Mo Joint



- Braze penetration to several hundred micrometers in 5 min.
- No effect of fiber ply orientation on infiltration.
- Improved wetting by Ti in braze facilitated infiltration.
- No reaction choking and flow cessation from carbide forming reactions.
- Extensive infiltration of C-C consistent with sessile-drop tests (complete disappearance of drops in porous carbon!).

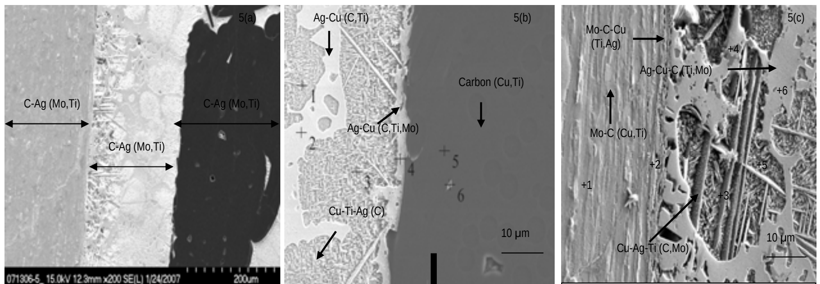
3-D C-C composite/Cusil ABA/Cu-clad-Mo joint



- High concentrations of Ti at interface.
- Two-phase eutectic structure in braze (Ag-rich light-grey and Cu-rich dark grey).
- No melting of clad layer (M.P. of Cu: 1086ºC)
- Possible formation of titaium carbide ($Ti+C \rightarrow TiC$, ΔG = -171.18 kJ at 850°C).
- Sub-stoichiometric carbides ($TiC_{0.95}$, $TiC_{0.91}$, $TiC_{0.80}$, $TiC_{0.70}$, $TiC_{0.60}$ and $TiC_{0.48}$) may also form.

Location	C	Ti	Cu	Mo	Ag
Point 1	96.468	0.693	0.000	0.723	2.116
Point 2	35.131	49.912	5.203	0.941	8.813
Point 3	0.675	0.328	3.881	0.281	94.835
Point 4	0.000	2.437	89.469	0.000	8.094

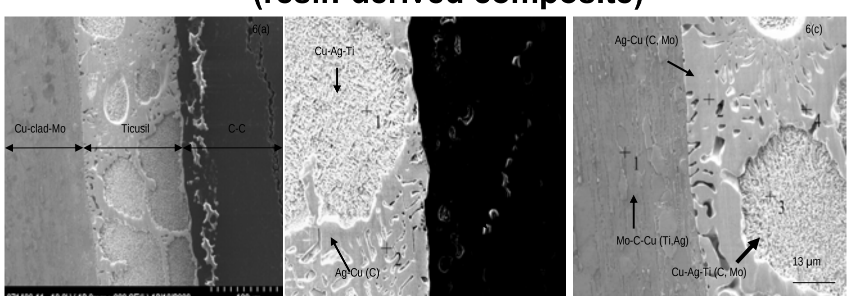
3-D C-C composite/Ticusil/Cu-clad-Mo joint



- **Some dissolution of carbon in braze (possibly due to higher temperature of Ticusil).**
- **Carbon also detected within the Cu-clad-Mo region.**

Location	C	Ti	Cu	Mo	Ag
Point 1	5.345	8.090	81.097	0.314	5.154
Point 2	2.097	0.228	4.540	0.508	92.627
Point 3	0.000	0.368	75.623	0.236	14.057
Point 4	1.852	10.084	2.776	0.382	88.623
Point 5	98.060	0.264	1.183	0.231	0.262
Point 6	26.237	54.484	16.697	0.553	2.030

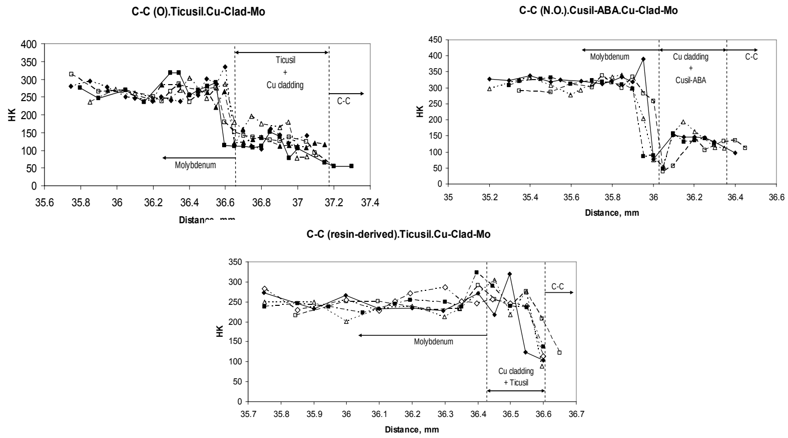
C-C composite/Ticusil/Cu-clad-Mo joint (resin-derived composite)



- **Cracks in composite (low inter-laminar shear strength).**
- **Braze displays eutectic structure with Ag- and Cu-rich phases.**
- **Precipitation of Ag-rich phase on C-C and Cu-clad-Mo surfaces.**
- **A small amount of Cu detected within the C-C composite.**

Location	C	Ti	Cu	Mo	Ag
Point 1	0.799	6.603	77.559	0.422	14.617
Point 2	2.198	0.495	9.874	0.460	86.973
Point 3	99.472	0.112	0.000	0.198	0.218
Point 4	78.303	0.527	18.766	1.136	1.268

Knoop Hardness across Brazed Joints

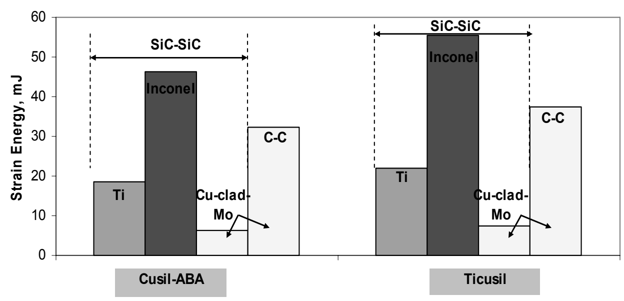


- **No effect of fiber ply orientation and composite type (CVI *vs* resin-derived) on HK within the braze region.**
- **HK of Mo substrate is ~200-330; HK depends on braze type: Ticusil (4.5%Ti) has higher HK (~85-200) than Cusil-ABA (1.75%Ti) (~50-150).**
- **Some effect of larger residual stresses in Ticusil because of its higher joining temperature.**

Strain Energy in Brazed Joints

Large strain energy → Greater tendency for fracture

(J.-W. Park, P. F. Mendez and T. W. Eagar, Acta Mater., 2002, 50(5), 883-899)



- **Less strain energy develops in Cusil-ABA joints than in Ticusil joints.**
- **Strain energy C-C/Ticusil/Cu-clad-Mo joints is 37.5 mJ.**
- **Ductile braze and Cu cladding prevented failure.**
- **Lower strain energy in comparable joints of Cu-clad-Mo with SiC-SiC.**

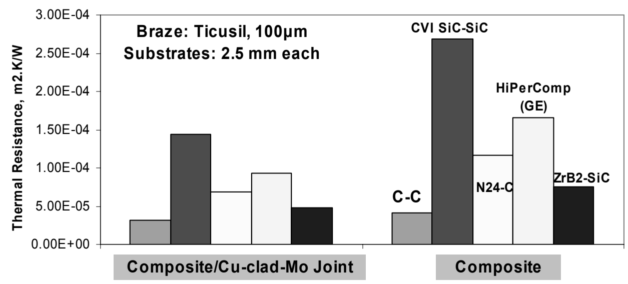
National Aeronautics and Space Administration

Estimation of Thermal Resistance in Brazed Joints

Effective thermal resistance (1-D steady-state conduction)

$$R_{eff} = \Sigma(\Delta x_i/K_i)$$

(Δx_i: thickness K_i: thermal conductivity)



- C-C/Cu-clad-Mo joints have 22% lower thermal resistance than C-C.
- There is some weight penalty in joining C-C to Cu-clad-Mo (39% increase in density).
- Potential benefit to join C-C to Cu-clad-Mo in thermal management systems.

National Aeronautics and Space Administration

Concluding Remarks

- C-C composites with CVI and resin-derived matrices were brazed to Cu-clad-Mo using active braze alloys.
- SEM and EDS revealed sound bonding and Ti segregation at interface and no evidence of extensive chemical attack of C-C. There was limited redistribution of alloying elements.
- De-lamination in resin-derived C-C was observed due to its low inter-laminar shear strength (ILSS). Extensive braze infiltration of inter-fiber channels occurred in 3D composites.
- Sharp hardness gradients occurred at Cu-clad-Mo/braze interface. Ticusil exhibited greater hardness (~85-250 HK) than Cusil-ABA (~50-150 HK). This may be due to higher Ti content of Ticusil (4.5% Ti) than Cusil-ABA (1.75% Ti).
- C-C/Cu-clad-Mo joints may have ~22% lower thermal resistance compared to C-C composites.

Acknowledgement

- **Ceramics Branch, NASA Glenn Research Center, Cleveland, OH for support to R. Asthana.**