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Diffusion bonding of Cu to Nb

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U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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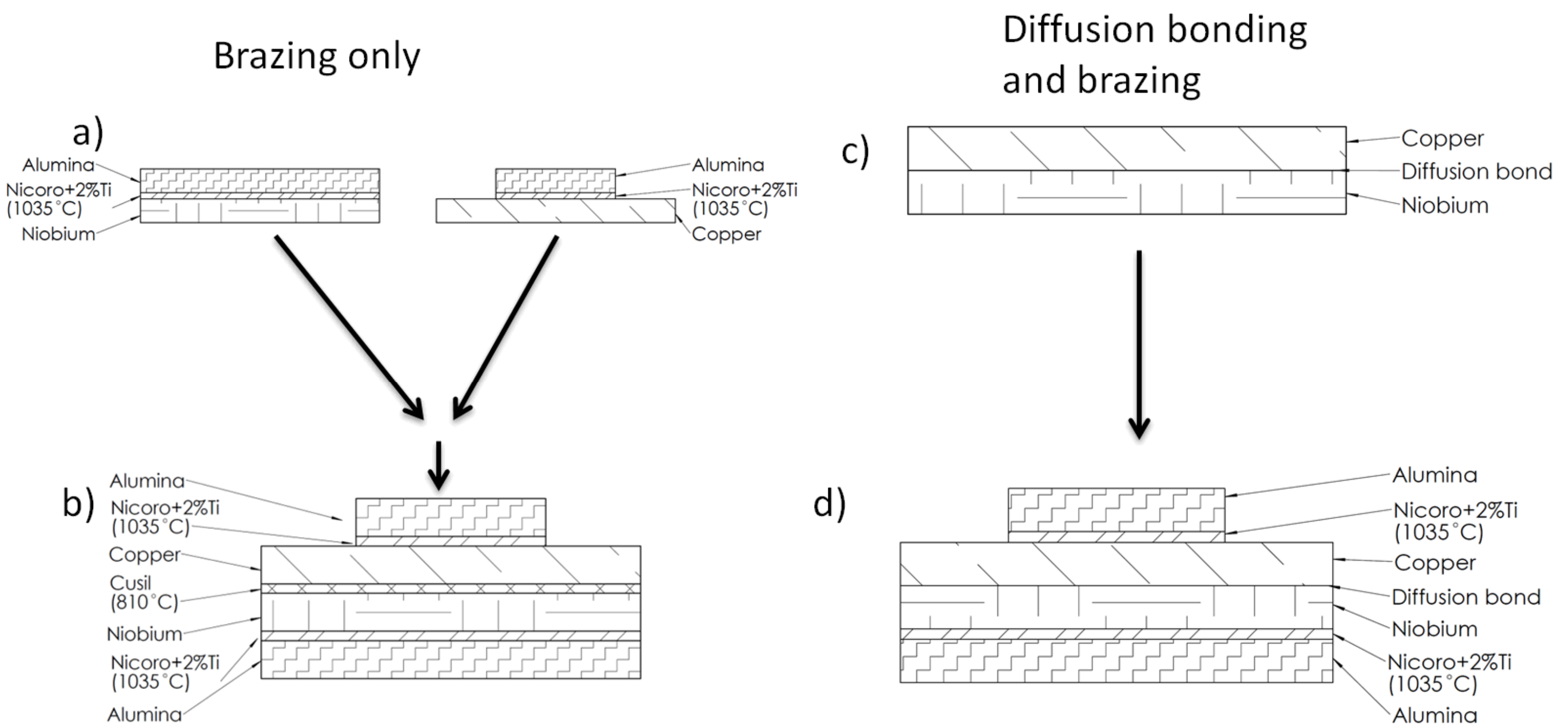
Outline

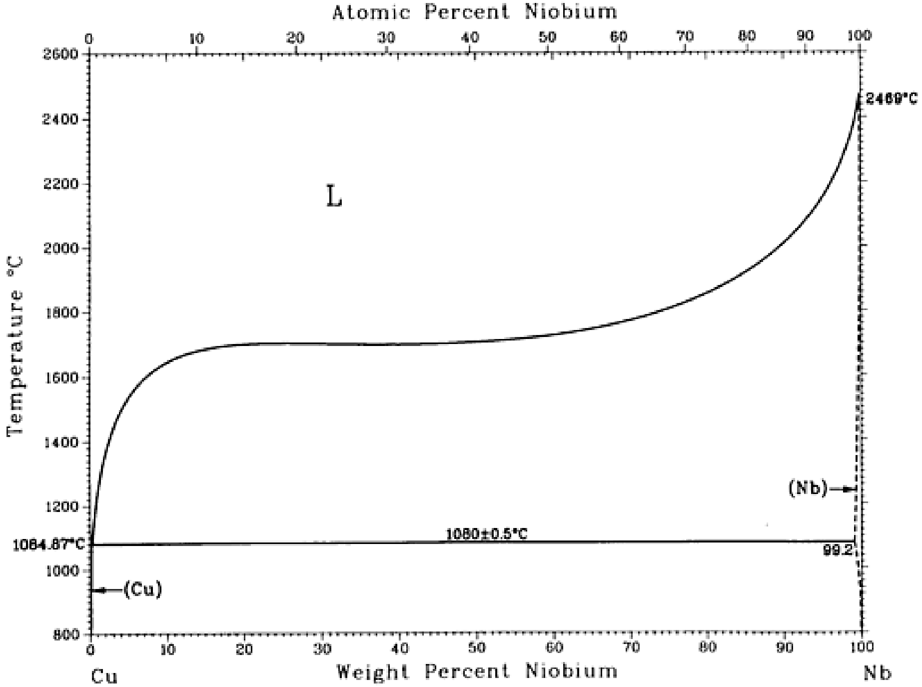
- Introduction
- Why Cu and Nb?
  - Material properties
  - Analysis of Cu and Nb
- Diffusion bonding research
- What is diffusion bonding?
  - Diffusion bonding parameters
- Experimental procedure
- Preliminary results
- Conclusions
- Planned work

Introduction

- Join two ceramic components
- Current method multiple braze cycles
  - Alumina to Nb, Cu to Nb, Cu to alumina
- Goal: Join two ceramic components
  with an optimized diffusion bonded
  Cu-Nb substrates

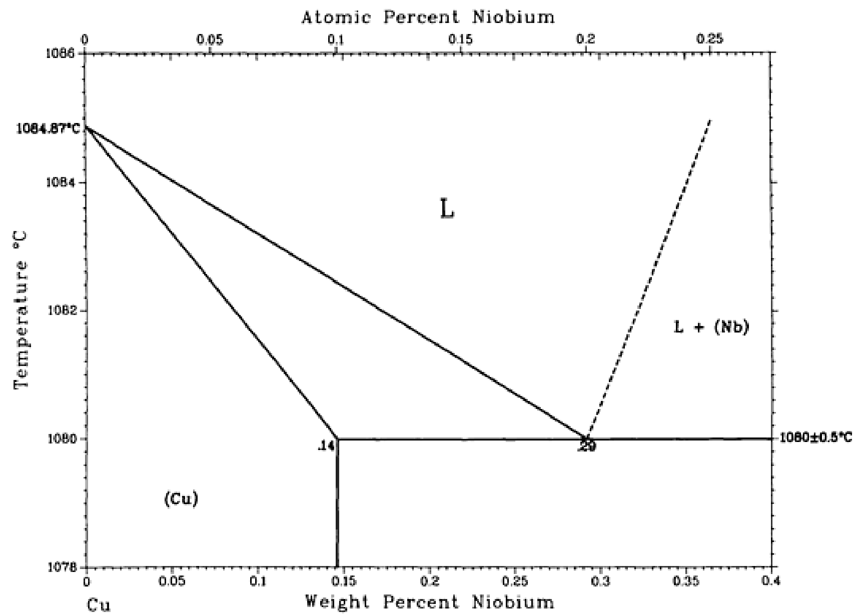


Why Cu and Nb?



Cu-Nb phase diagram

Advantages
-Braze ability
-No intermetallic formation



Enlargement of the Cu-rich portion of the Cu-Nb system.

Difficulties
-Limited solubility
-Different melting temperatures

T. Massalski, Binary alloy phase diagrams, 1, 938-939, (1986)

Properties: Cu and Nb

Crystal structure and physical properties for Cu and Nb [15, 16]

	Cu	Nb
Crystal structure	FCC	BCC
Atomic radius, [pm]	128	146
Cell parameter 'a', [pm]	361.49	330.04
Space-group	Fm-3m	Im-3m
CTE, [µm/m/K]	16.5×10^{-6}	7.3×10^{-6}
Melting point, [ºC]	1084	2469

$T_m(Cu) << 0.5\ T_m(Nb)$

Analysis of Cu and Nb

- The material used in this work is:
  - Copper-OFHC-hard (127µm thick (0.005 in)): ASTM-B152, C10200
  - Niobium (508µm thick (0.020 in)): ASTM-B393, R04210, Type 2

Lot analysis of Cu and Nb [PPM]

	Cu	Nb
O		10
P	20	
C		10
N		10
H		0.3
Fe	7	10
Ni	2	10
Si		10
W		30
Mo		10
Ta		600
Zr		20
Co	3	
Pb	1	
Ag	10	

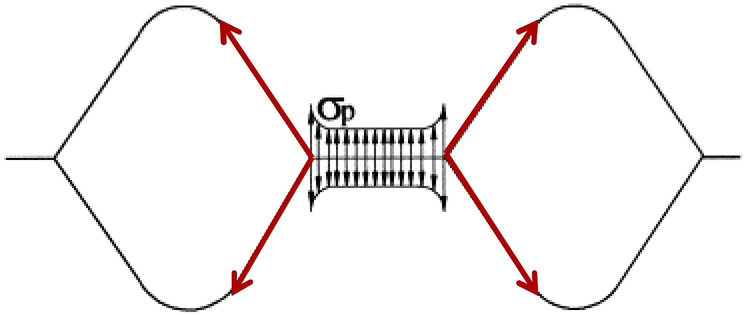
Diffusion bonding research

- Diffusion bonding of Cu to Nb research is insufficient
- Research has been conducted on:
  - Cu-W with bond aid[1,2]
  - Cu-Alumina[3,4,5,6,7]
  - Nb-Alumina[3]
  - Cu to 304L stainless steel[8]
  - Affects of annealing on Cu-Nb nanolayered thin films[17,18,19,20,21,22]
    - Kurdjumov-Sachs (K-S) orientation relation

      $(111)_{fcc} \parallel (110)_{bcc}$ and $\langle 110\rangle_{fcc} \parallel \langle 111\rangle_{bcc}$

What is diffusion bonding?

- Solid state joining of similar or dissimilar components
- 2 sets of mechanisms during diffusion bonding
  - Deformation mechanisms
  - Diffusion mechanisms



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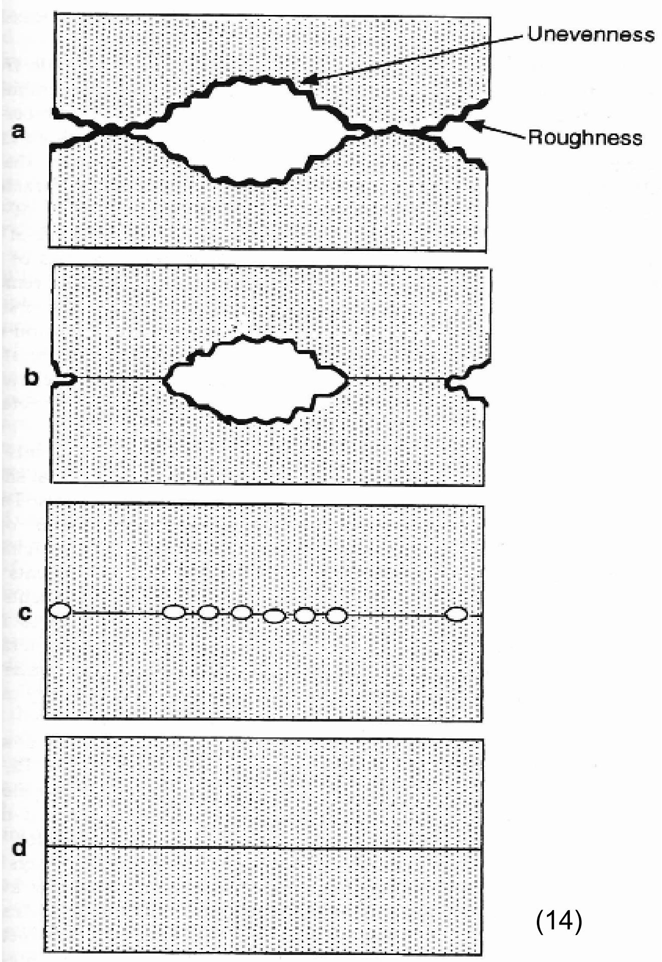

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Maxwell relation* $\left(\frac{\partial \mu}{\partial p}\right)_{T,N} = \left(\frac{\partial V}{\partial N}\right)_{T,p}$

Derived from Gibbs free energy eq. $\mu = -\sigma dT + \nu dp; \frac{S}{N} = \sigma, \frac{V}{N} = \nu$

*K. Stowe, *Introduction to statistical mechanics and thermodynamics*, John Wiley and sons, New York, 209-231, (1984)

Diffusion-controlled creep

-Diffusion-controlled creep is the dominate diffusion mechanism

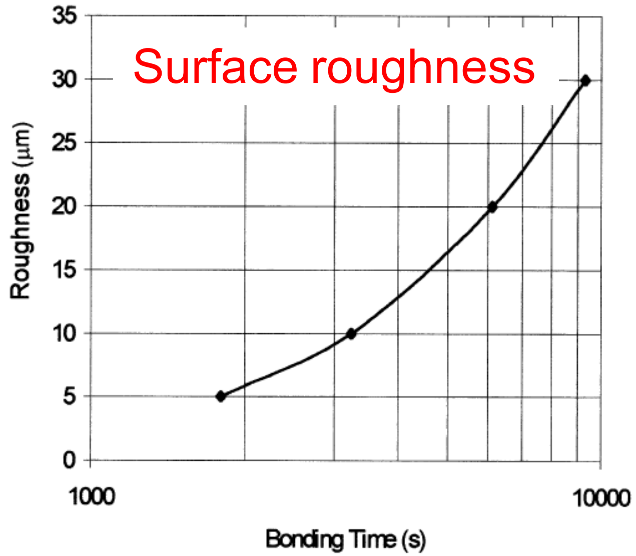
$$Nabarro - Herring\ creep: \dot{\varepsilon}_{NH} = A_{NH} D_v \frac{\Omega\sigma}{\kappa T d^2}$$

$$Coble\ creep: \dot{\varepsilon}_C = A_C D_{GB(S)} \frac{\Omega\sigma\delta}{\kappa T d^3}$$

$\dot{\varepsilon}_{NH}$: Creep strain rate for Nabarro-Herring creep
$\dot{\varepsilon}_C$: Creep strain rate for Coble creep
$A_{c,NH}$: Coble creep, and Nabarro-Herring constants
$D_{v,GB(S)}$: Volume and grain boundary diffusivities
Ω: Atomic volume
K: Boltzman's constant (1.38×10^{-23} J K^{-1})
T: Absolute temperature
D: Grain size
δ: Grain boundary width
σ: Applied stress

Diffusion bonding parameters

- Surface preparation
  - Surface roughness
  - Cleanliness
- Temperature
- Pressure
- Time



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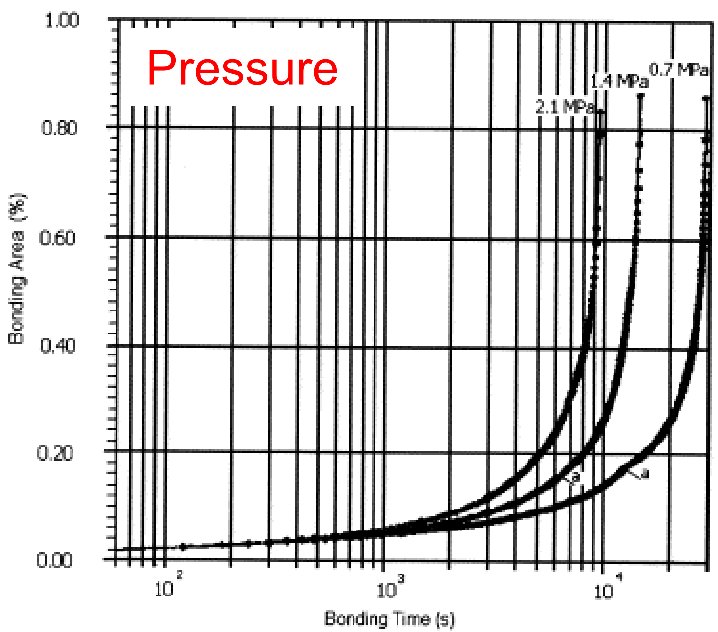

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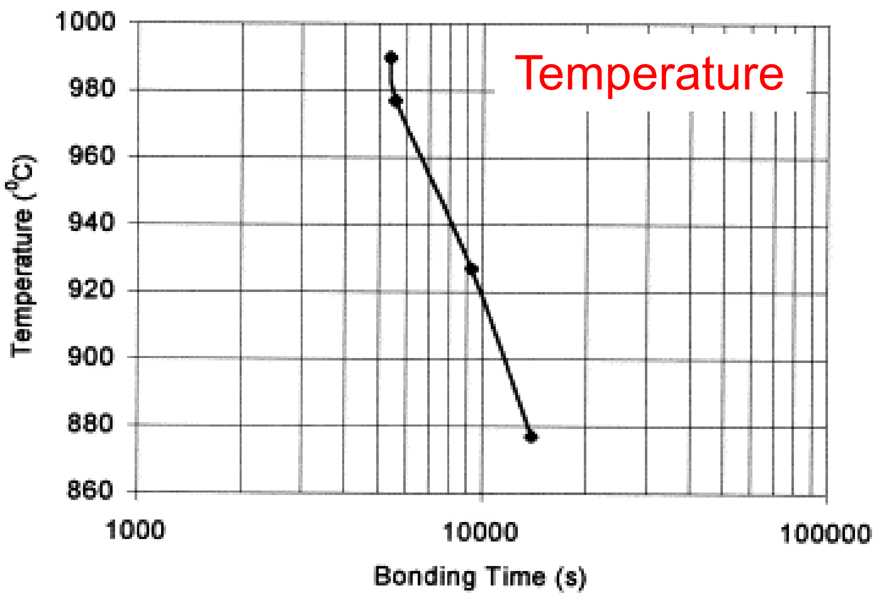

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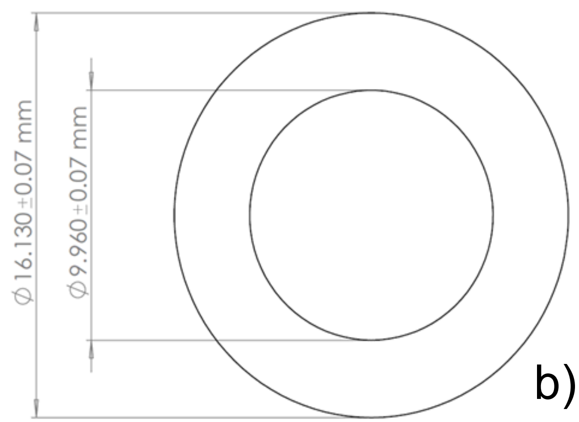
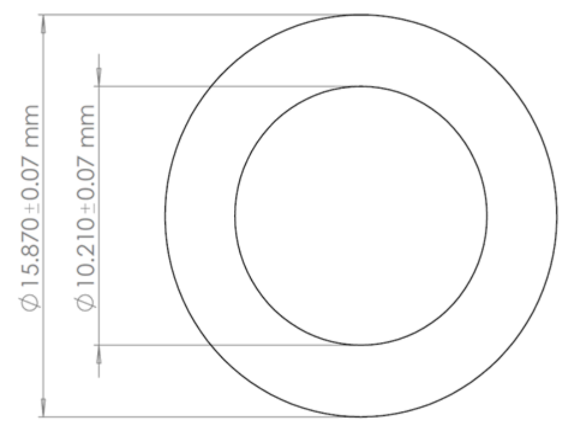
Sample preparation for diffusion bonding and brazing

- Diffusion bonding
  - Cut samples
  - Manually polish
  - Electropolish Cu and Nb
  - Vacuum bake Nb
- Brazing
  - a) machine Cu-Nb washers from bonded substrates
  - Bake Cu-Nb substrates
  - b) Punch Nicoro®+2%Ti braze washers



a)

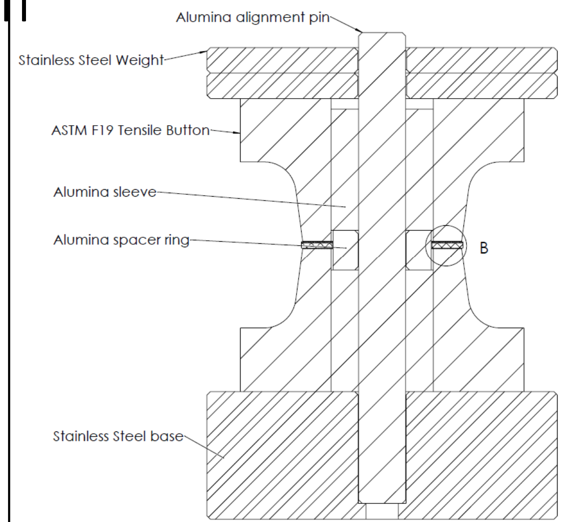


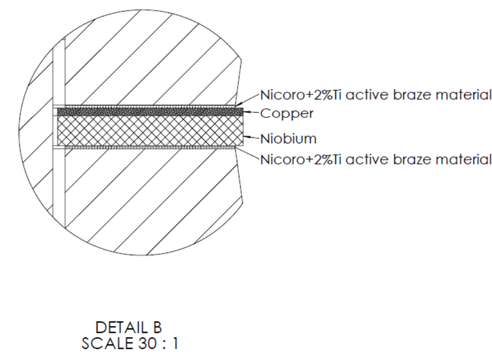
b)

Sample analysis for diffusion bonding and brazing

- Braze samples with Nicoro®+2%Ti
- Helium leak check with
  He mass spec leak detector
- SEM, EDS, and TEM
- Mechanical testing
  - Tensile test
  - Peel test

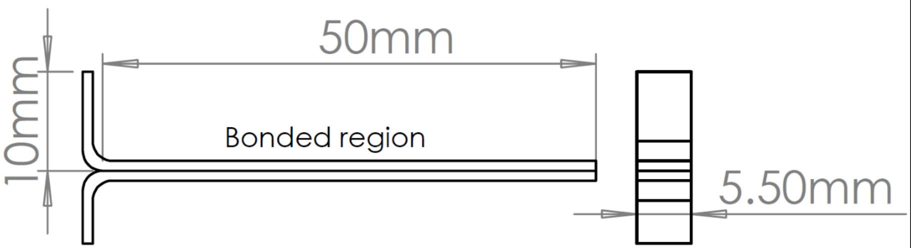
Tensile button sample
-Tests braze assembly



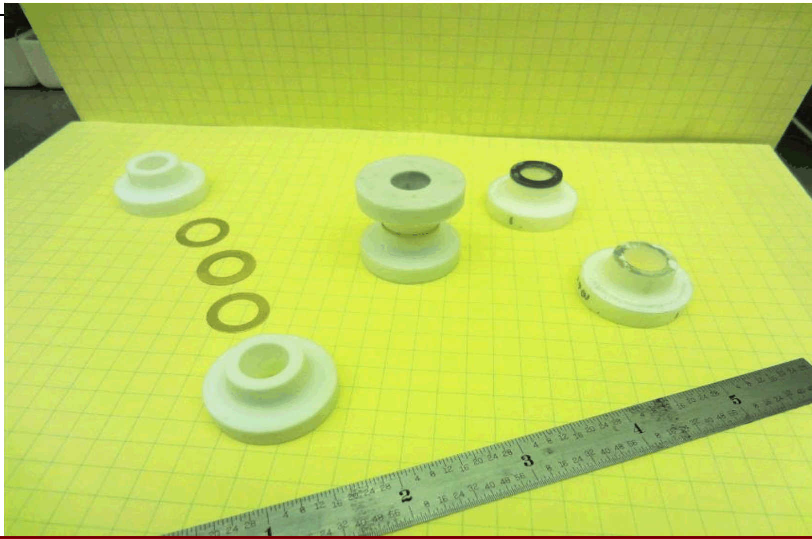


DETAIL B
SCALE 30 : 1

Peel test sample



-Tests diffusion bonded interface



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Preliminary experiments

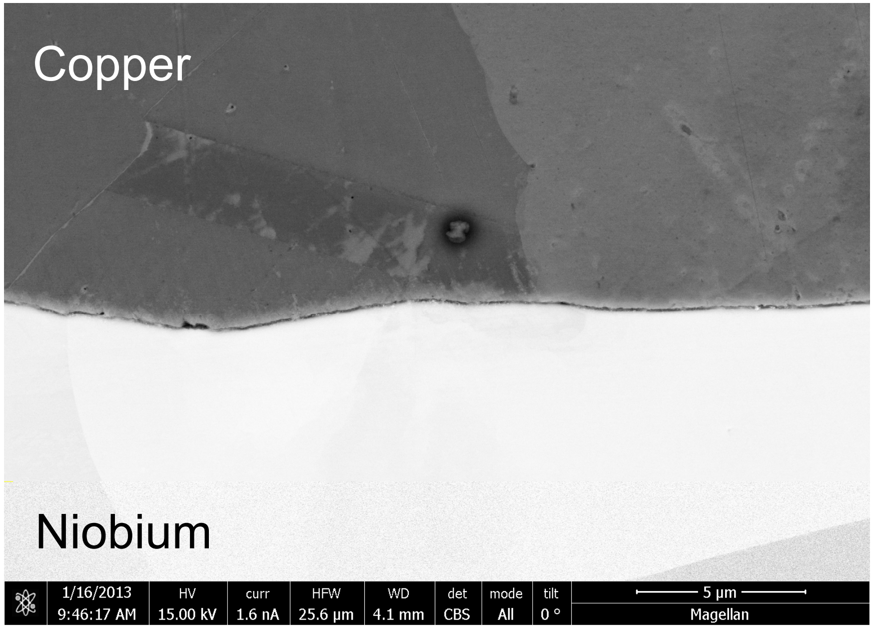
- Preliminary results for experiments in yellow

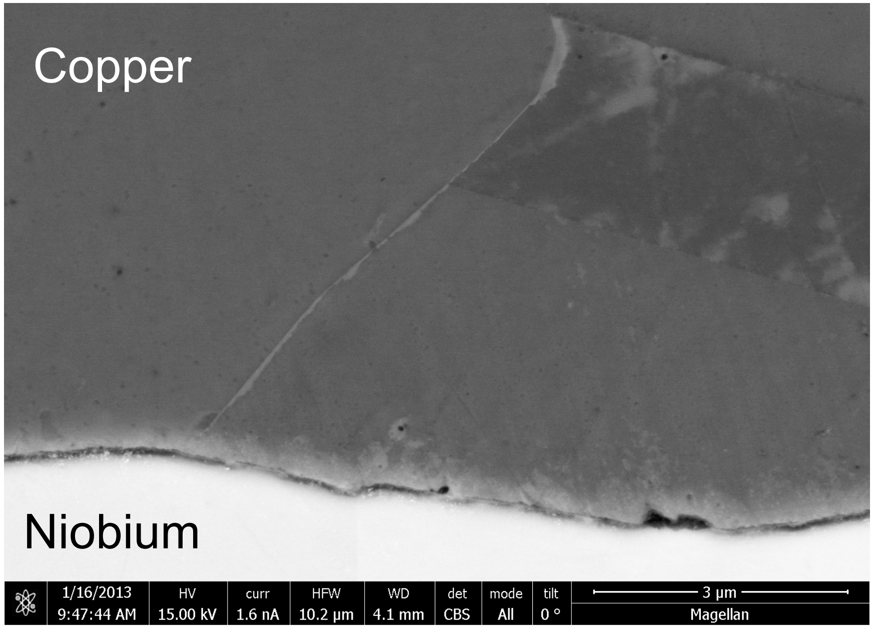
	Temperature [^{o}C]	Pressure [Mpa (KSI)]	Time [Hours]	Material thickness		Load applied [^{o}C]
				Cu [µm]	Nb [µm]	
	950	10.3 (1.5)	0.5	127	508	950
	950	4.1 (0.6)	3	127	508	950
	1000	6.9 (1.0)	1.5	127	508	1000
Sample 1	1035	4.1 (0.6)	3	50.8	1016	1035
	1035	4.1 (0.6)	3	127	508	1035
Sample 2	1050	4.1 (0.6)	3	50.8	254	1050
	1050	4.1 (0.6)	3	127	508	1050
	1050	10.3 (1.5)	0.5	127	508	1050

Sample 1: SEM, and tensile buttons
Sample 2: SEM

Results: SEM analysis of sample 1

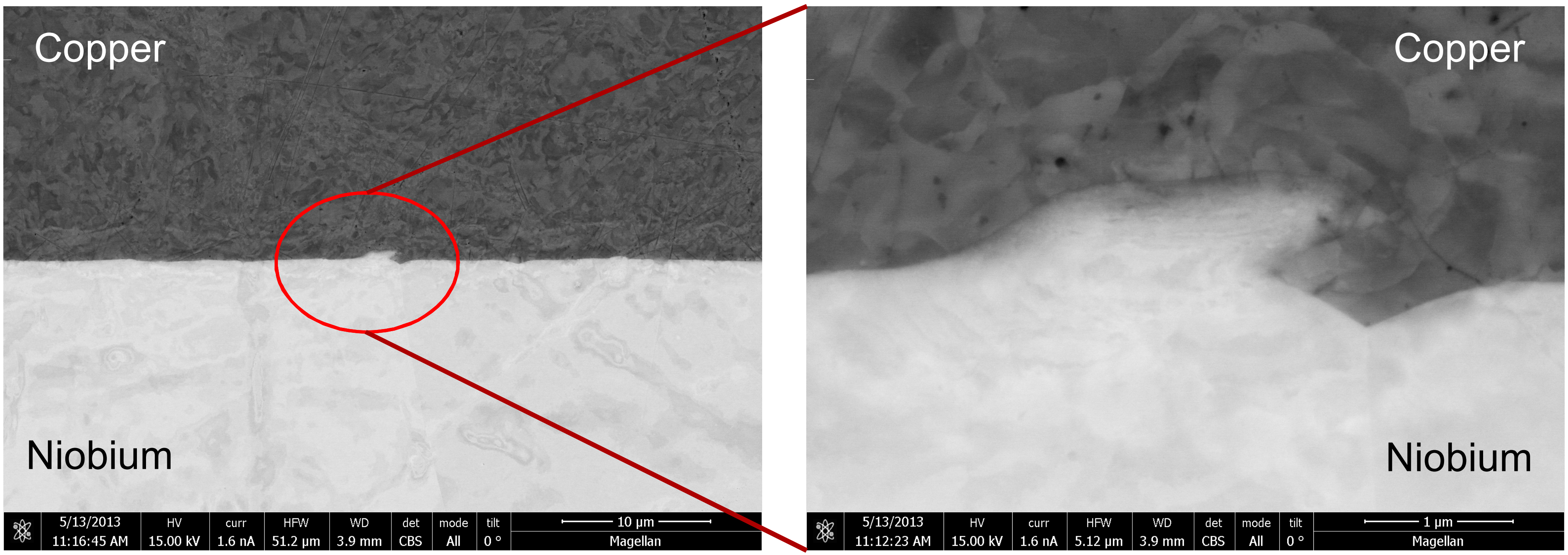
- Cu-Nb diffusion bonded interface.
- Bonding parameters: 1035°C, 3 hours, 4MPa (0.6 KSI), <2x10^{-6}Torr





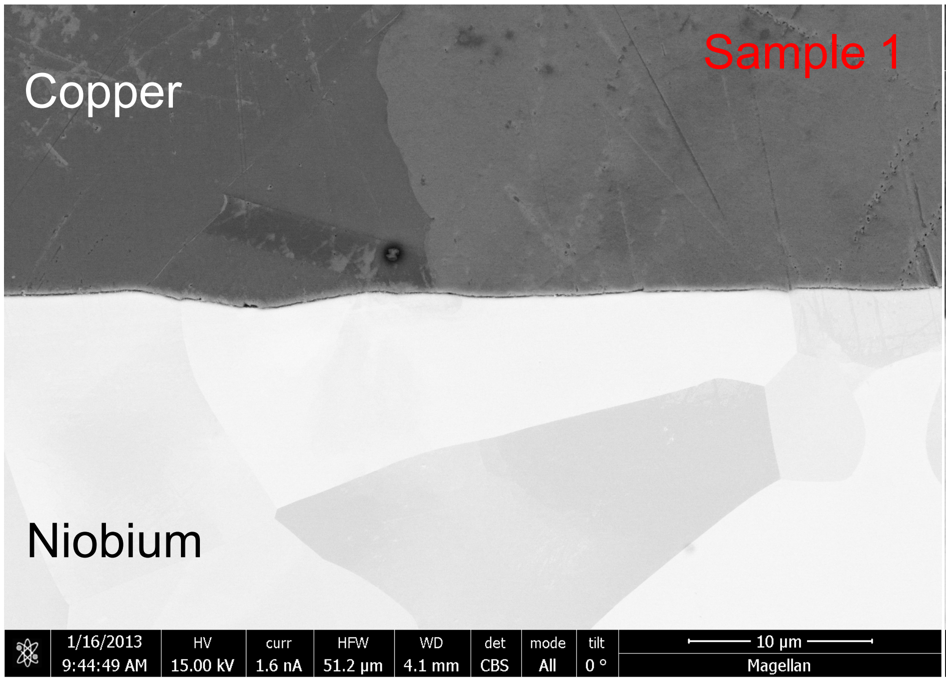
Results: SEM analysis of sample 2

- Cu-Nb diffusion bonded interface.
- Bonding parameters: 1050°C, 3 hours, 4MPa (0.6KSI), <2x10^{-6}Torr

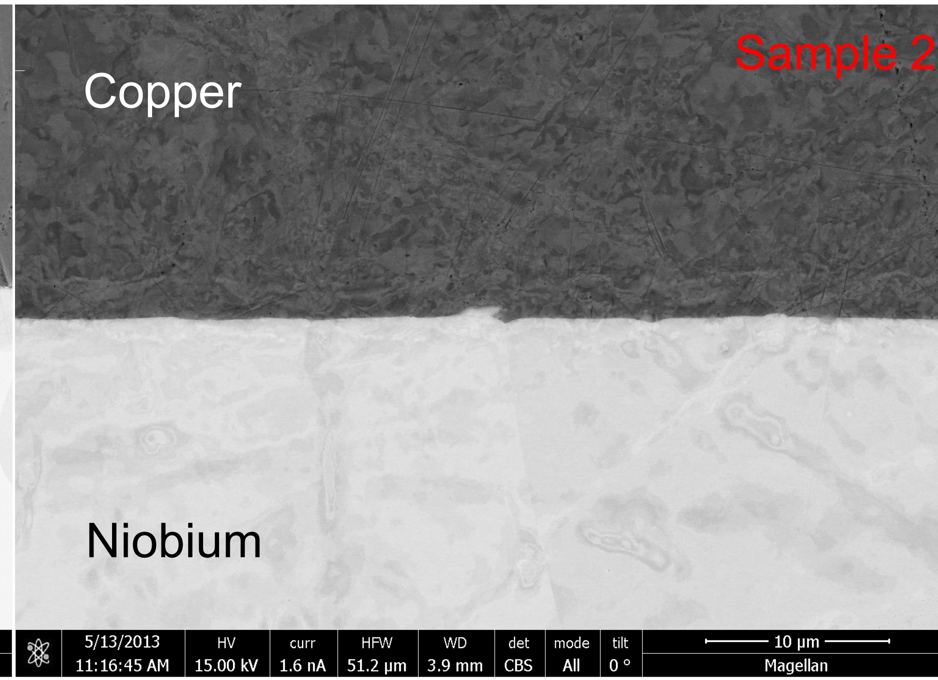


Results: Interface comparison

Bonding temperature: 1035^{o}C



Bonding temperature: 1050^{o}C



Constant bonding parameters: 3 hours, 4MPa (0.6KSI), <2x10^{-6}Torr

Cu grain size difference was observed:

Sample 1: >10µm

Sample 2: <1µm

Mechanical testing results

- Mechanical testing was performed on diffusion bonded sample 1
- Tensile button test
  - Diffusion bonding parameters: 1035°C, 3 hours, 4MPa (0.6 KSI)
  - Brazing profile: 15°C/min to 900°C, hold 10.0 mins; 10°C/min to 1035°C, hold 5.0m mins; 25C/min to RT, <2x10^{-6} Torr.

Sample	Leak rate [sccs]		Load [lbs]	KSI*	MPa		Failure Location
1-1	NDL		1602.40	8.91	61.42		Braze joint between ceramic and Nb
1-2	NDL		2797.00	15.55	107.21		Ceramic on Cu side
1-3	NDL		2142.10	11.91	82.11		Ceramic on Cu side

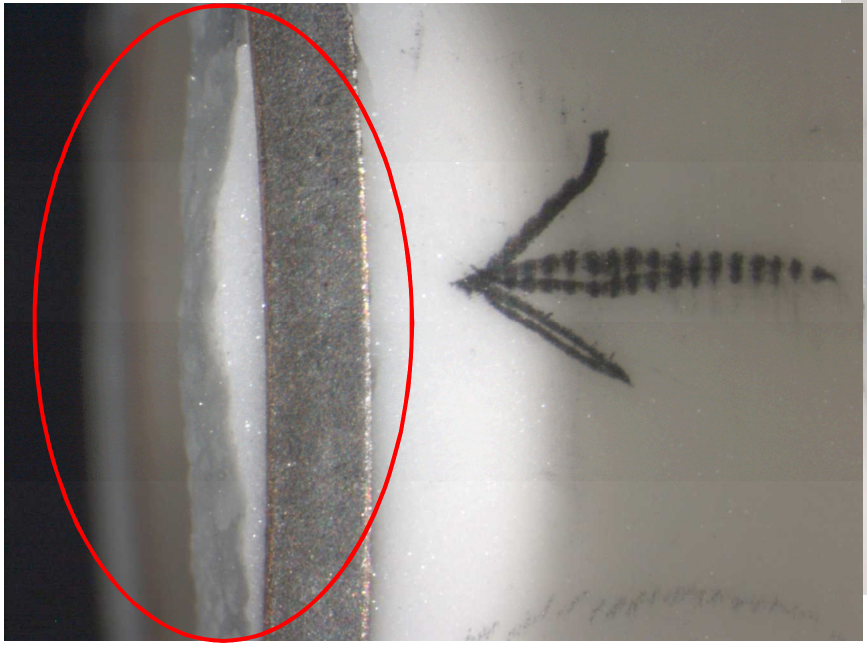
Avg.:	12.12	83.58

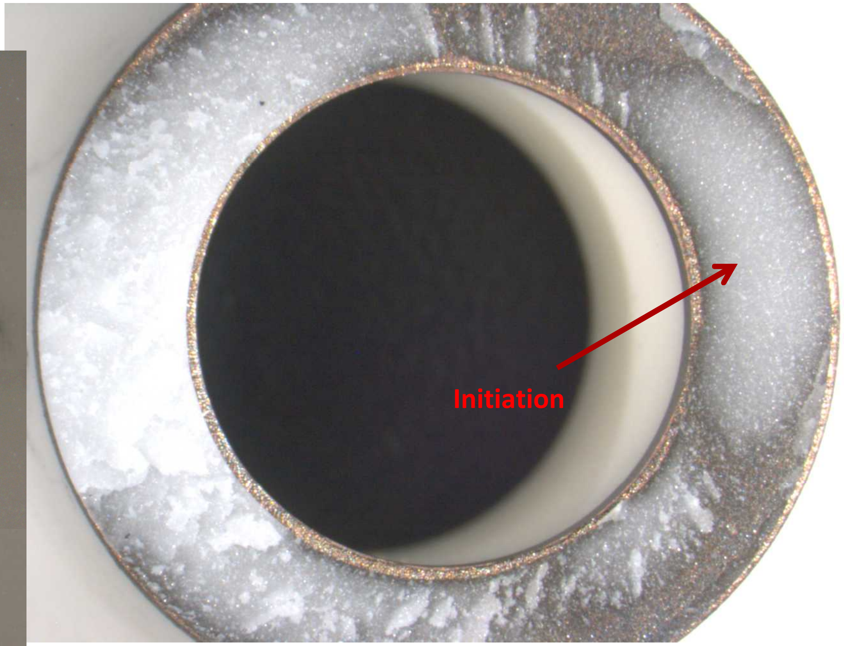
*Surface area: 0.179873 sqr in

Results: Fracture of tensile button

- Diffusion bonding parameters: 1035°C, 3 hours, 4MPa (0.6KSI)
- Sample tensile strength 107.21 MPa (15.5 KSI)

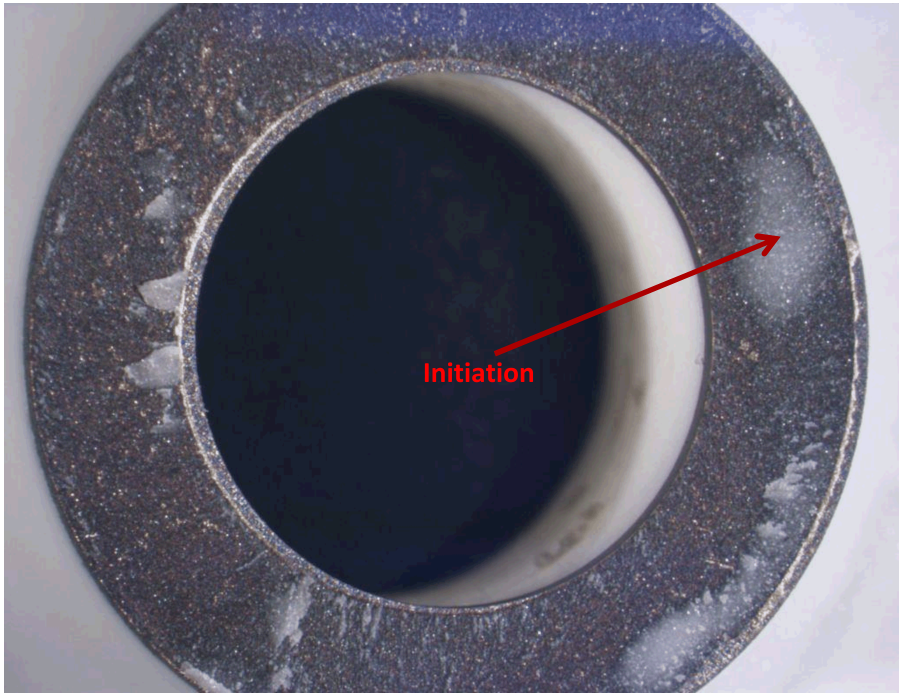
Sample 1-2





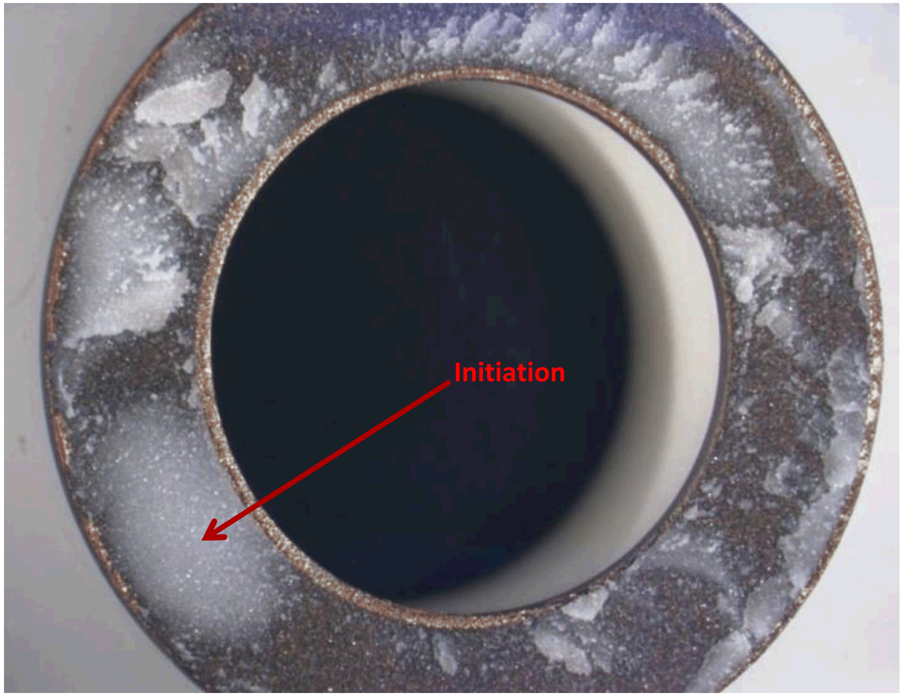
Results: Fracture of tensile button

Sample 1-1



Tensile strength: 61.42 MPa
(8.91 KSI)

Sample 1-3

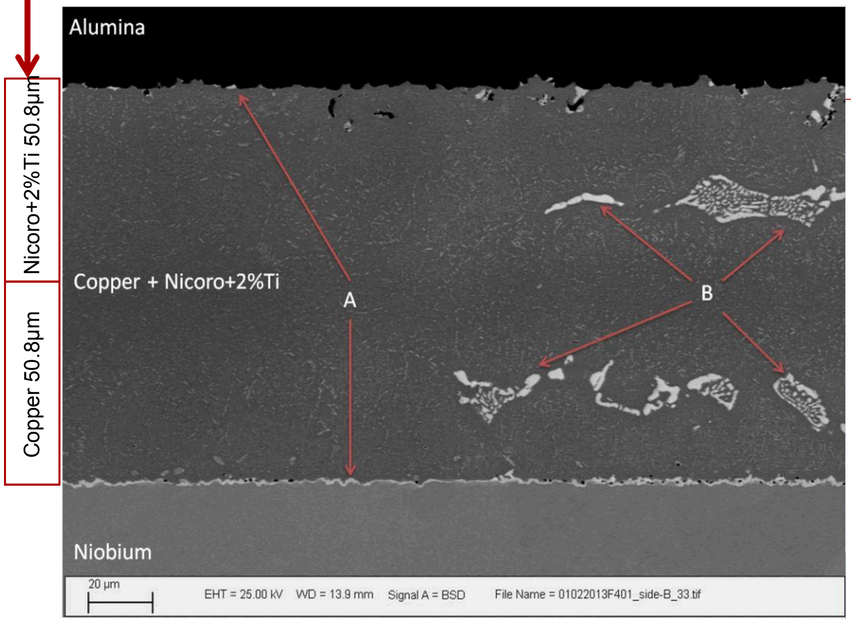


Tensile strength: 82.11 MPa
(11.91 KSI)

Results: Dissolution of Cu in braze joint

Original thickness

Cu-alumina braze joint after brazing



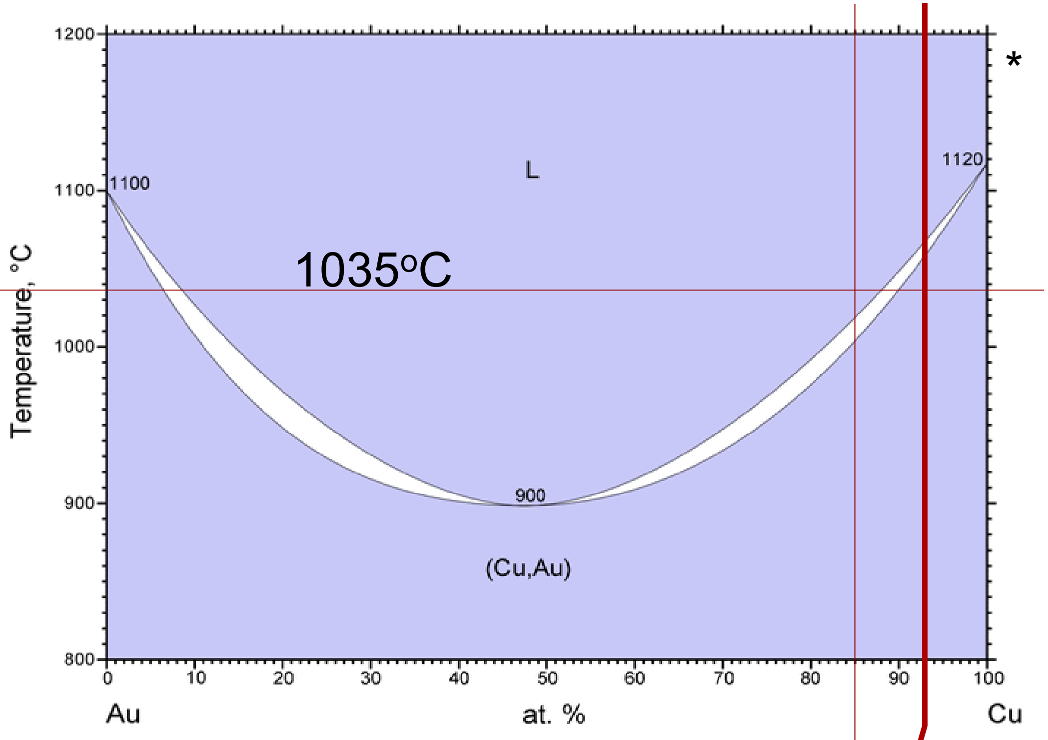
SEM of sample 1-2

Tensile strength: 107.21 Mpa

Original thicknesses;

Cu: 50.8μm, Nb: 1016μm

Nicoro®+2%Ti: 50.8μm



Initial composition of braze alloy 85 at %Cu

Final composition of braze joint 93 at %Cu

*W. T. Zheng, Study on phase diagrams for Ag-Au, Au-Cu binary alloys at high temperature, 36, 75-79, (1991)

Conclusions

- Diffusion bonding of Cu to Nb has been successfully demonstrated
- In order to improve the diffusion bond and decrease the amount of interfacial voids, the pressure needs to be increased when temperature is decreased, and the temperature needs to be increased when pressure is decreased.

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Planned Work: Design of Experiments

- Conduct remaining experiments

Temperature [ºC]	Pressure [Mpa (KSI)]	Time [Hours]	Material thickness		Load applied [ºC]
			Cu [µm]	Nb [µm]	
950	10.3 (1.5)	0.5	127	508	950
950	4.1 (0.6)	3	127	508	950
1000	6.9 (1.0)	1.5	127	508	1000
1035	4.1 (0.6)	3	50.8	1016	1035
1035	4.1 (0.6)	3	127	508	1035
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1050	4.1 (0.6)	3	127	508	1050
1050	10.3 (1.5)	0.5	127	508	1050

- 3 additional experiments to determine optimal parameters
- 2 final experiments to confirm optimal parameters

Planned Work: Analysis

- Braze Cu-Nb diffusion bonded samples to ASTM F19 alumina tensile buttons with the Nicoro®+2%Ti braze filler alloy.
- Characterize the diffusion bond and brazed joint strength by peel testing, and tensile testing.
- Examine diffusion bonded interfaces and brazed joints with SEM, EDS and TEM of metallographic samples, and optical microscopy.

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Questions and Comments