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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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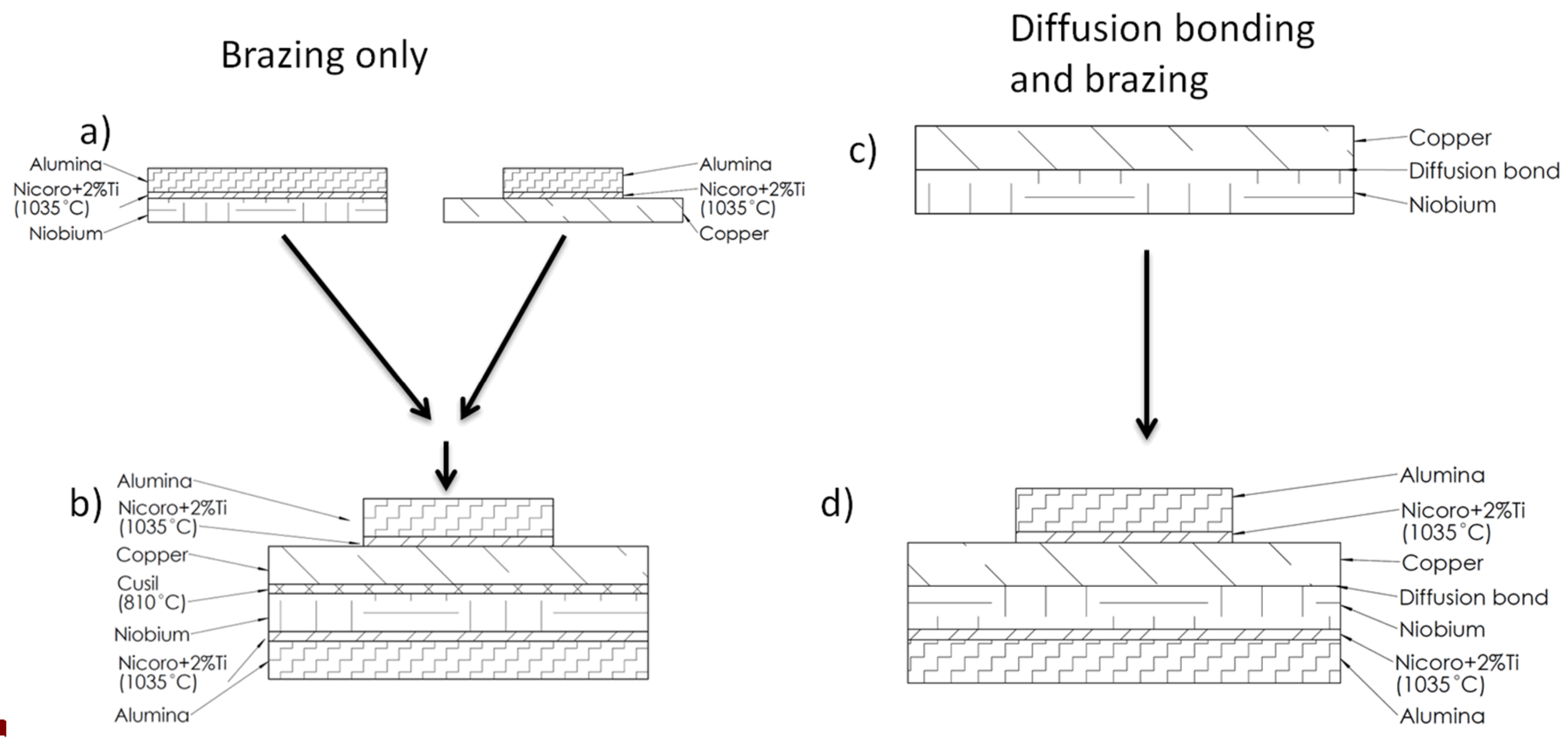
Outline

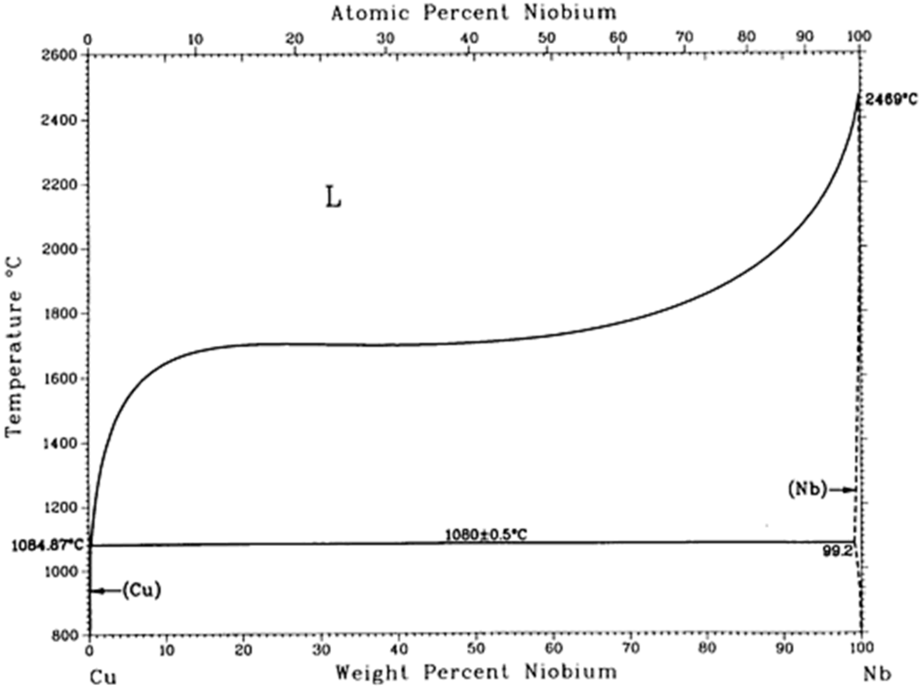
- Introduction
- Why Cu and Nb?
  - Material properties
- What is diffusion bonding?
  - Diffusion bonding parameters
- Experimental procedure
- Results
- Conclusions

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

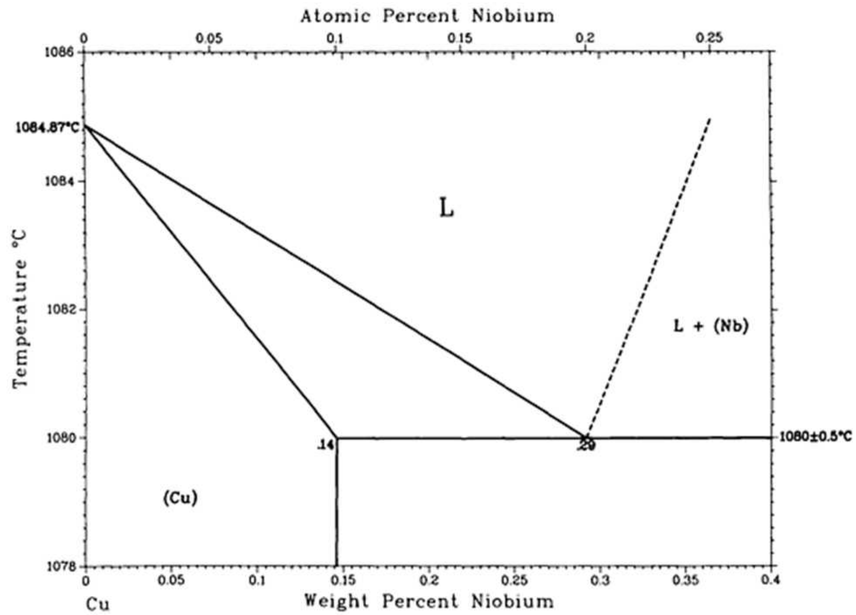


Why Cu and Nb?



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

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

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

What is diffusion bonding?

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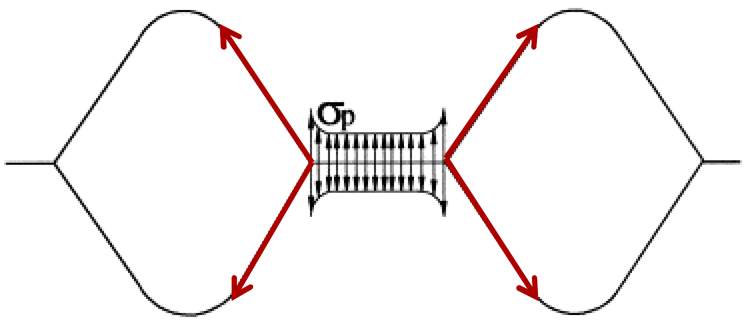
- Solid state joining of similar or dissimilar components
- 2 sets of mechanisms during diffusion bonding
  - Deformation mechanisms
  - Diffusion mechanisms



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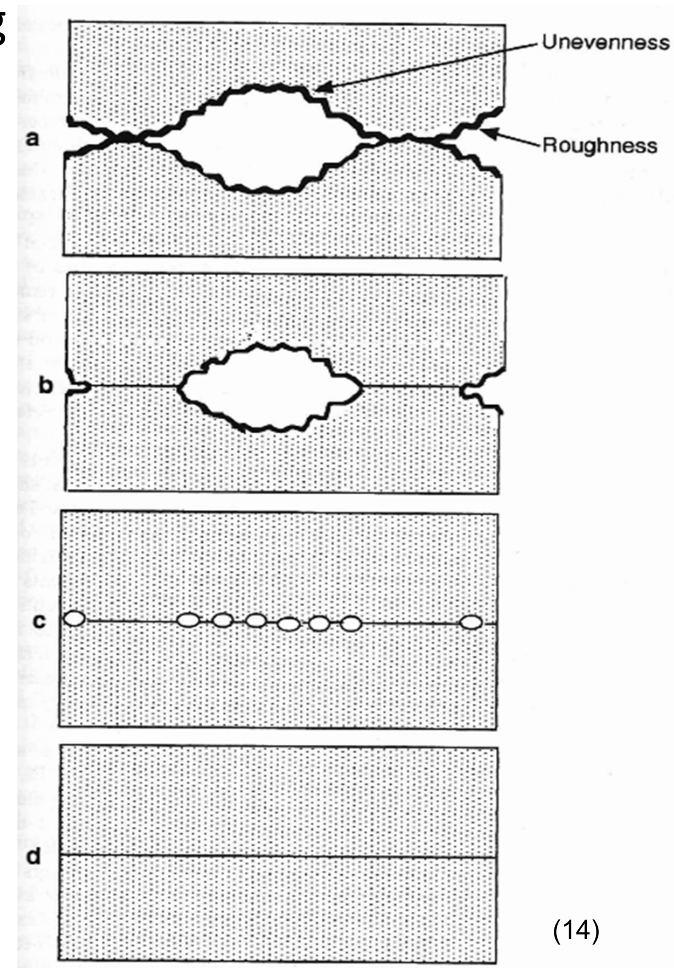

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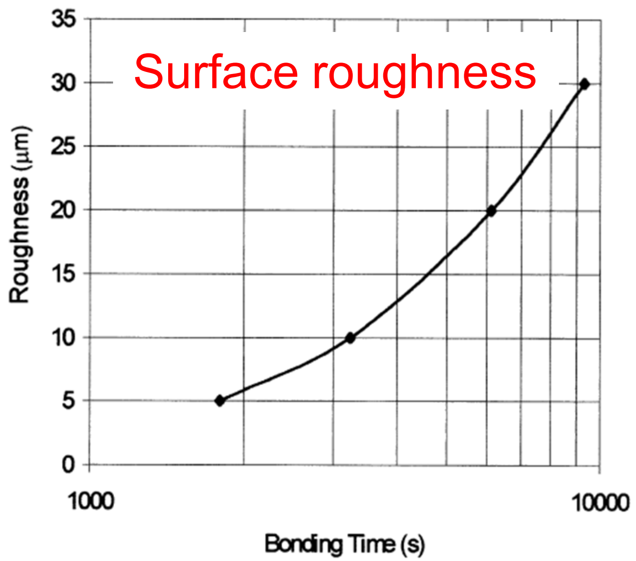
Maxwell relation* $$\left(\frac{\partial \mu}{\partial p}\right)_{T,N} = \left(\frac{\partial V}{\partial N}\right)_{T,p}$$

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 bonding parameters

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



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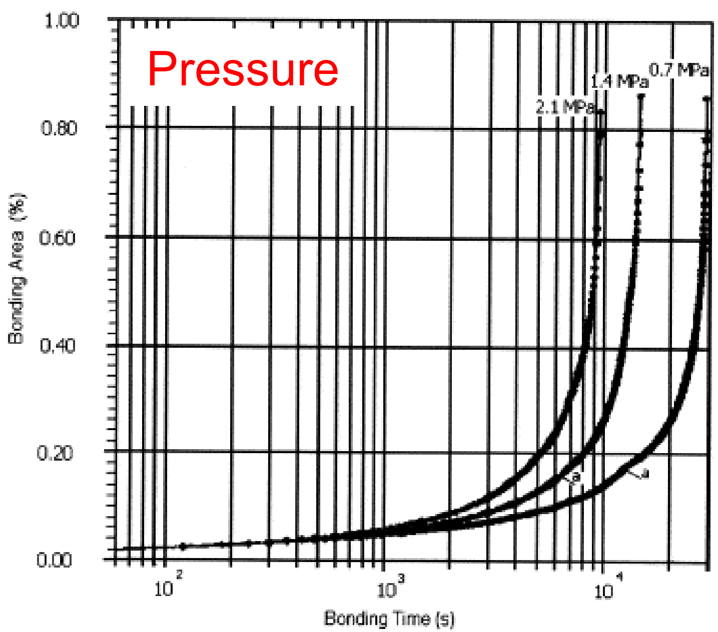

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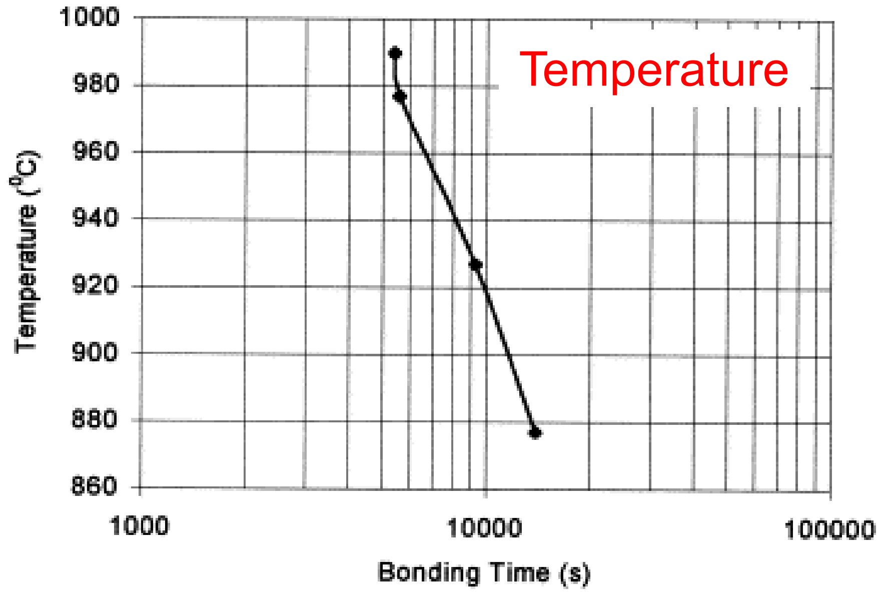

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  - Diffusion bonding parameters
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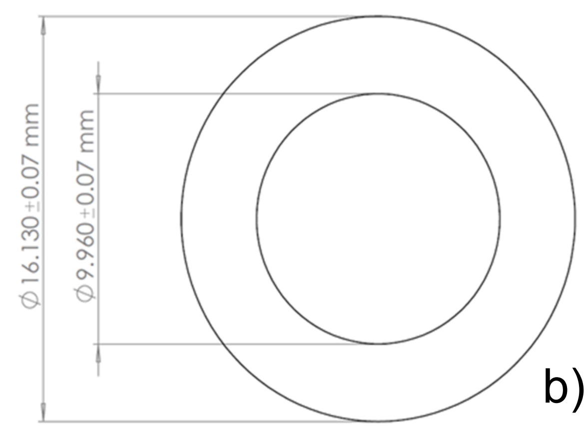
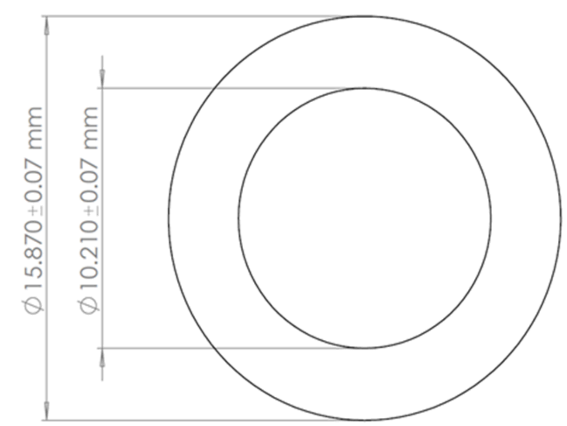
Sample preparation for diffusion bonding and brazing

- Diffusion bonding
  - Cut samples
  - Degrease samples in a Lenium vapor degreaser
  - Vacuum anneal Nb 1400°C
- Brazing
  - a) Wire EDM Cu-Nb washers from bonded substrates
  - Bake Cu-Nb substrates (800°C) to remove machining oils
  - b) Punch Nicoro®+2%Ti braze washers

a)

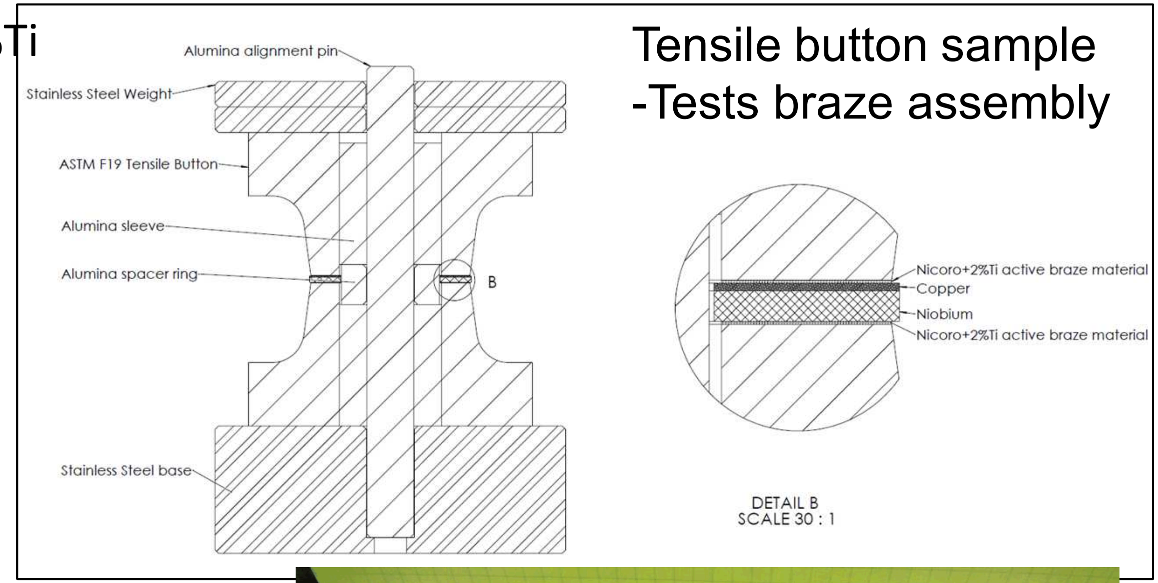


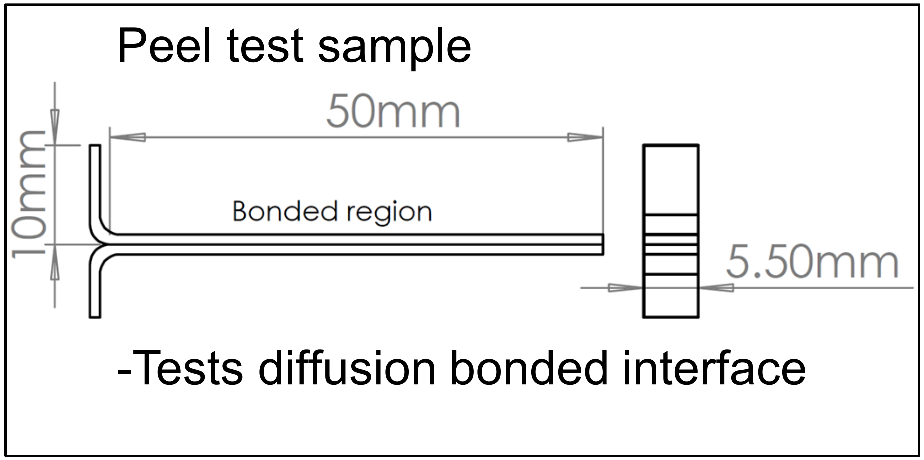
b)

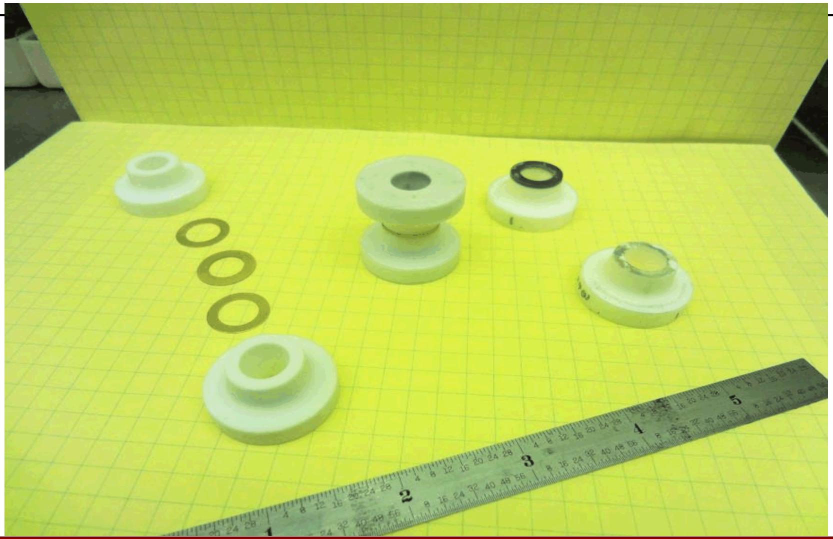


Sample analysis for diffusion bonding and brazing

- 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







Experiments

- Experiments conducted to determine best bonding parameters

	Temperature [°C]	Pressure [Mpa (KSI)]	Time [Hours]	Material thickness		Load applied [°C]
				Cu [μm]	Nb [μm]	
Run 1	950	10.3 (1.5)	0.5	127	508	950
Run 2	950	4.1 (0.6)	3	127	508	950
Run 3	1000	6.9 (1.0)	1.5	127	508	1000
Sample 1	1035	4.1 (0.6)	3	50.8	1016	1035
Run 4	1035	4.1 (0.6)	3	127	508	1035
Sample 2	1050	4.1 (0.6)	3	50.8	254	1050
Run 5	1050	4.1 (0.6)	3	127	508	1050
Run 6	1050	10.3 (1.5)	0.5	127	508	1050

Sample 1: SEM, and tensile buttons
Sample 2: SEM
Run 1,2,4: SEM
Run 3,5,6: SEM, and tensile buttons

Optimization experiments

- Experiments to determine optimal bonding parameters
  - Based of SEM results from initial experiments

	Temperature [°C]	Pressure [Mpa (KSI)]	Time [Hours]	Material thickness		Load applied [°C]
				Cu [µm]	Nb [µm]	
Run 7	1050	10.3 (1.5)	0.25	127	508	1050
Run 8	1050	4.1 (0.6)	1.5	127	508	1050
Run 9	1050	4.1 (0.6)	0.5	127	508	1050

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Experiments

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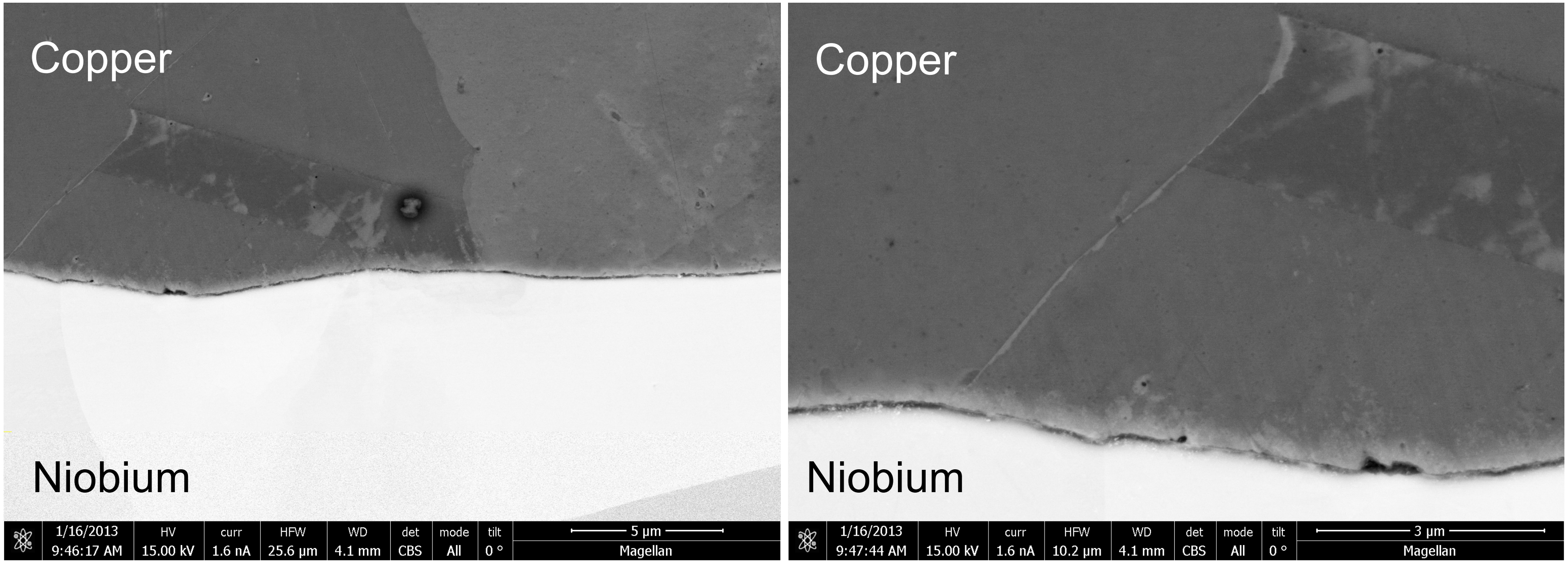
	Temperature [°C]	Pressure [Mpa (KSI)]	Time [Hours]	Material thickness		Load applied [°C]
				Cu [µm]	Nb [µm]	
Run 1	950	10.3 (1.5)	0.5	127	508	950
Run 2	950	4.1 (0.6)	3	127	508	950
Run 3	1000	6.9 (1.0)	1.5	127	508	1000
Sample 1	1035	4.1 (0.6)	3	50.8	1016	1035
Run 4	1035	4.1 (0.6)	3	127	508	1035
Sample 2	1050	4.1 (0.6)	3	50.8	254	1050
Run 5	1050	4.1 (0.6)	3	127	508	1050
Run 6	1050	10.3 (1.5)	0.5	127	508	1050

Sample 1: SEM, and tensile buttons
Sample 2: SEM
Run 1,2,4: SEM
Run 3,5,6: SEM, and tensile buttons

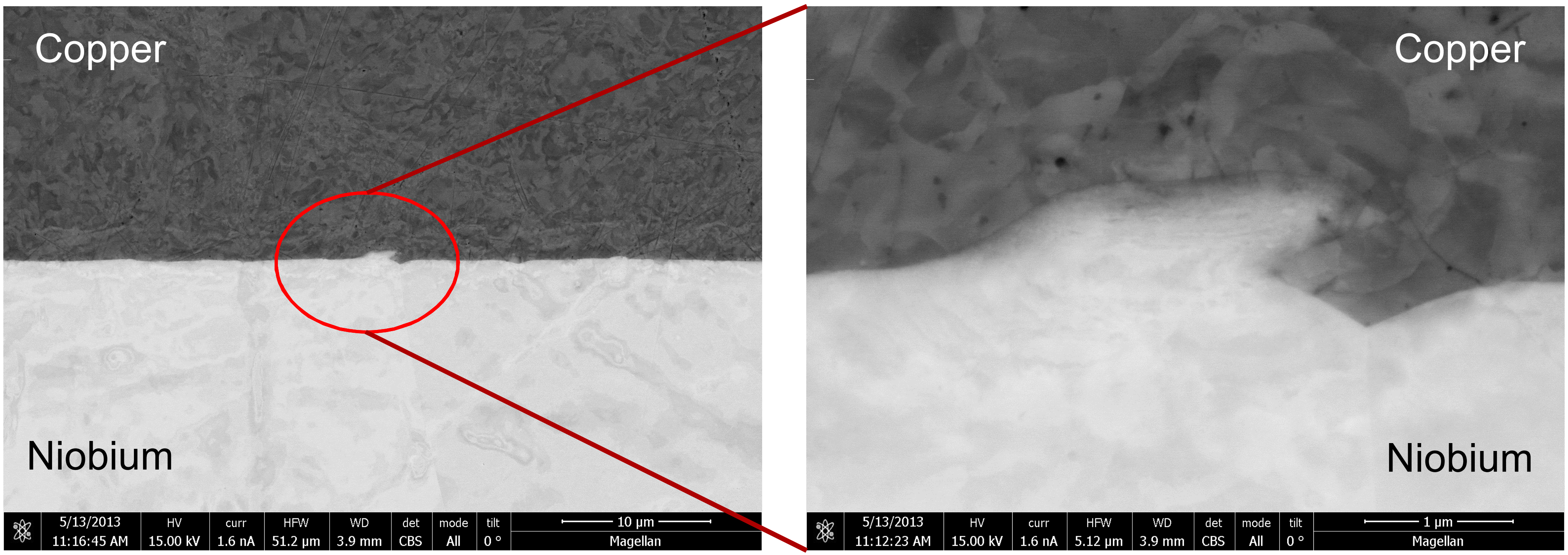
Results: SEM analysis of sample 1

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



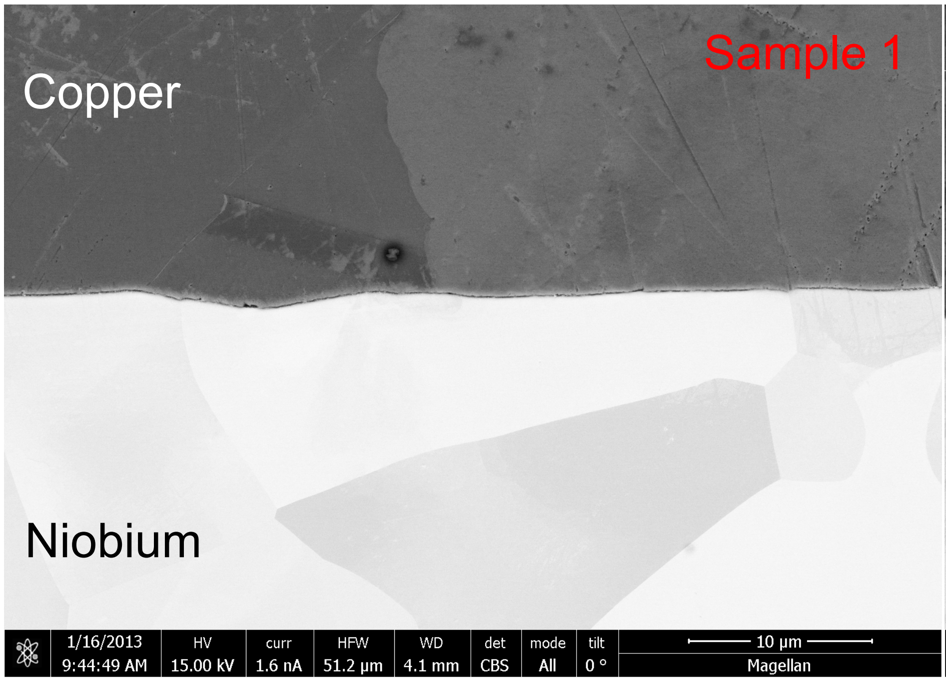
Results: SEM analysis of sample 2

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

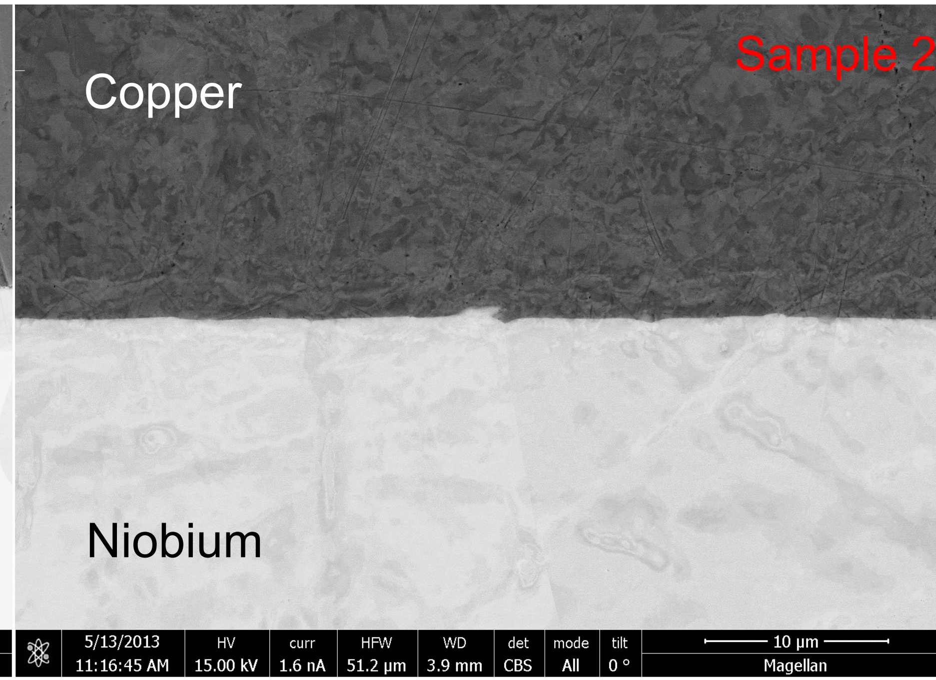


Results: Interface comparison

Bonding temperature: 1035^{o}C



Bonding temperature: 1050^{o}C



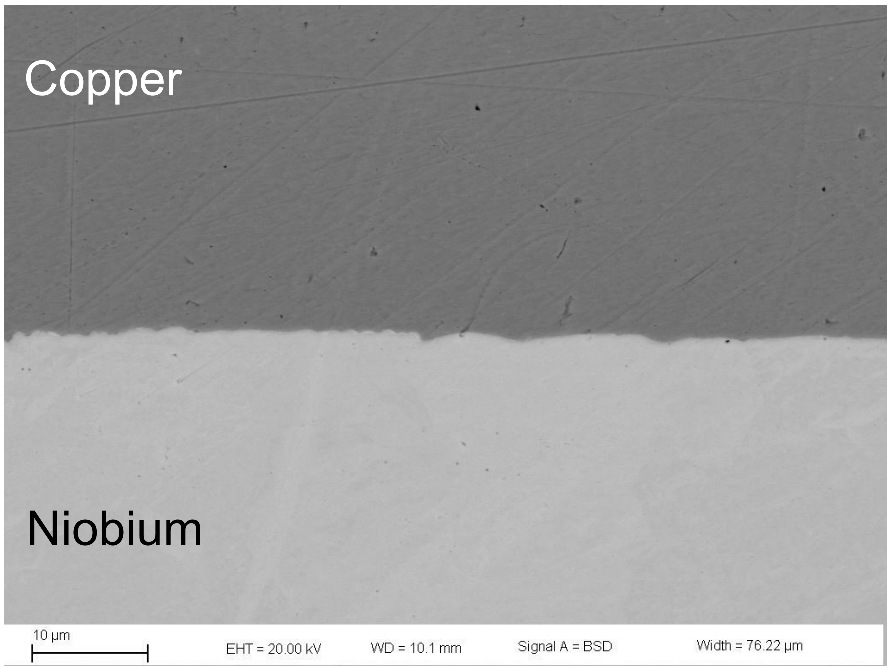
Constant bonding parameters: 3 hours, 4MPa (0.6KSI), $<2\times10^{-6}$Torr

Cu grain size difference was observed:

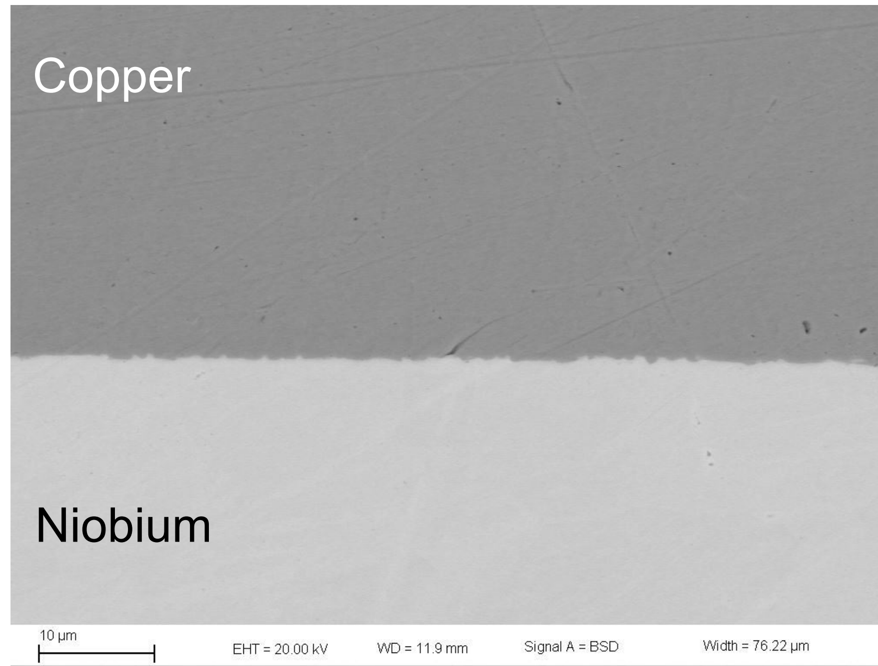
Sample 1: >10µm

Sample 2: <1µm

SEM analysis: Run 5 and 6

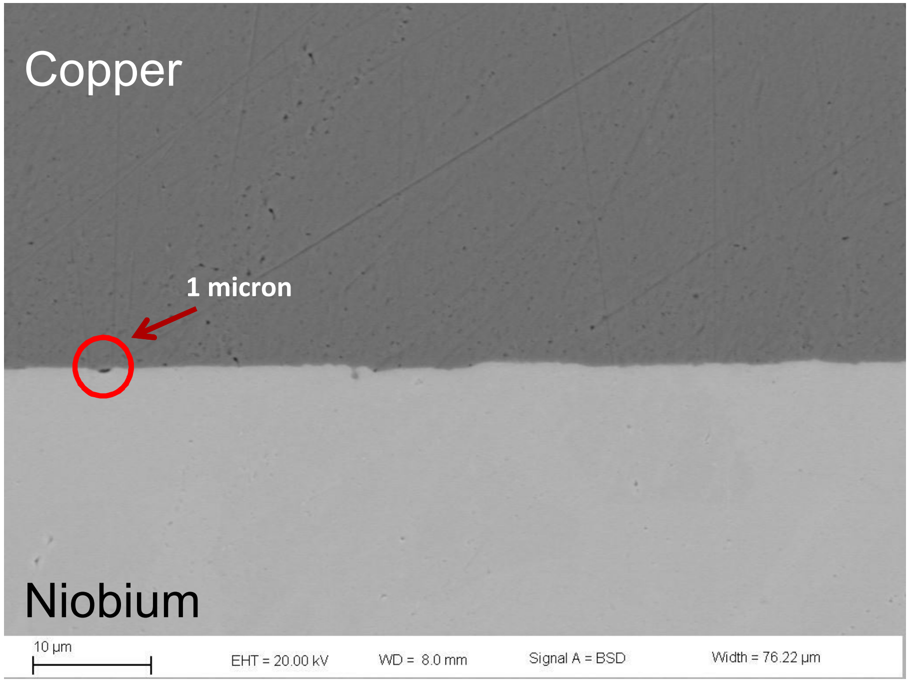


Run 5: 1050°C, 4.1 MPa, 3.0 hours

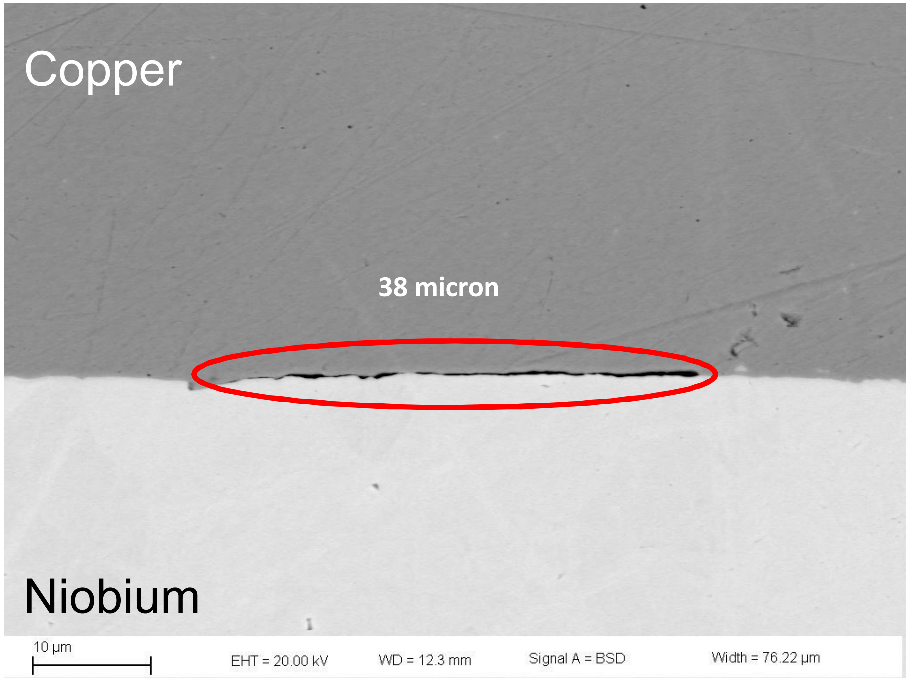


Run 6: 1050°C, 10.3 MPa, 0.5 hours

SEM analysis: Run 3 and 4

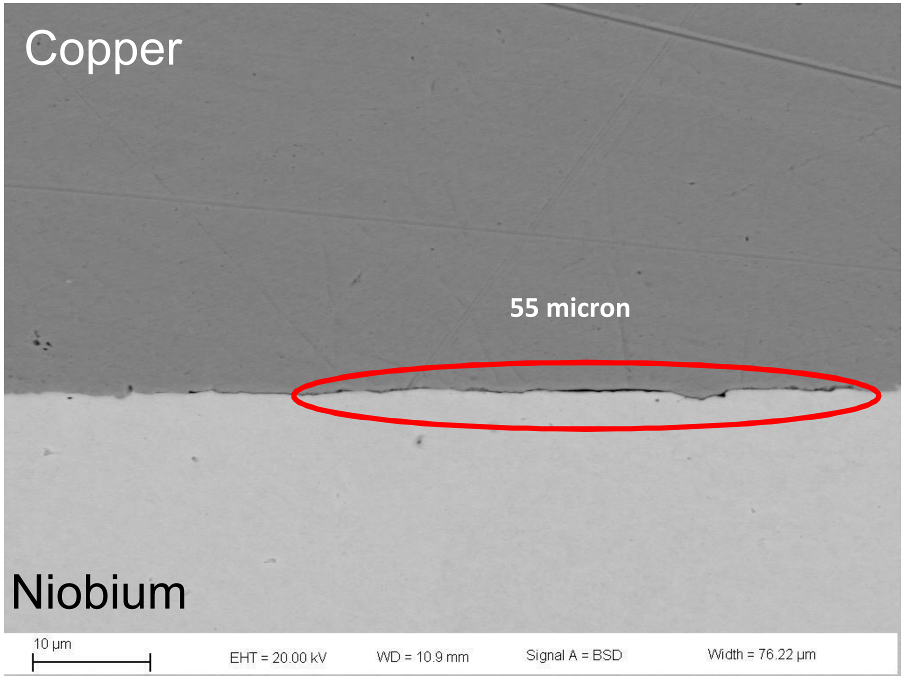


Run 3: 1000°C, 6.9 MPa, 1.5 hours

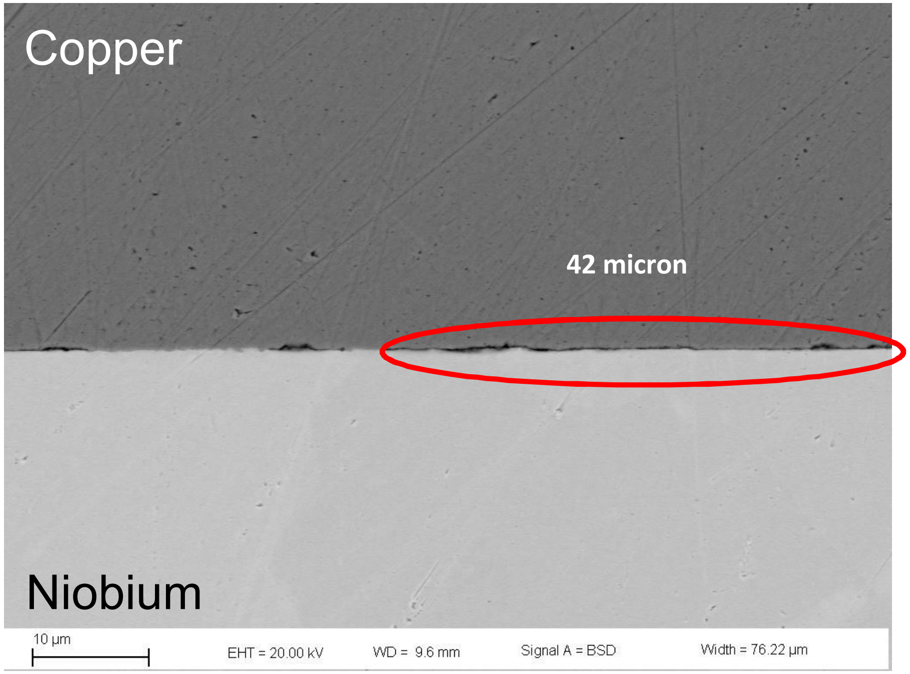


Run 4: 1035°C, 4.1 MPa, 3.0 hours

SEM analysis: Run 1 and 2

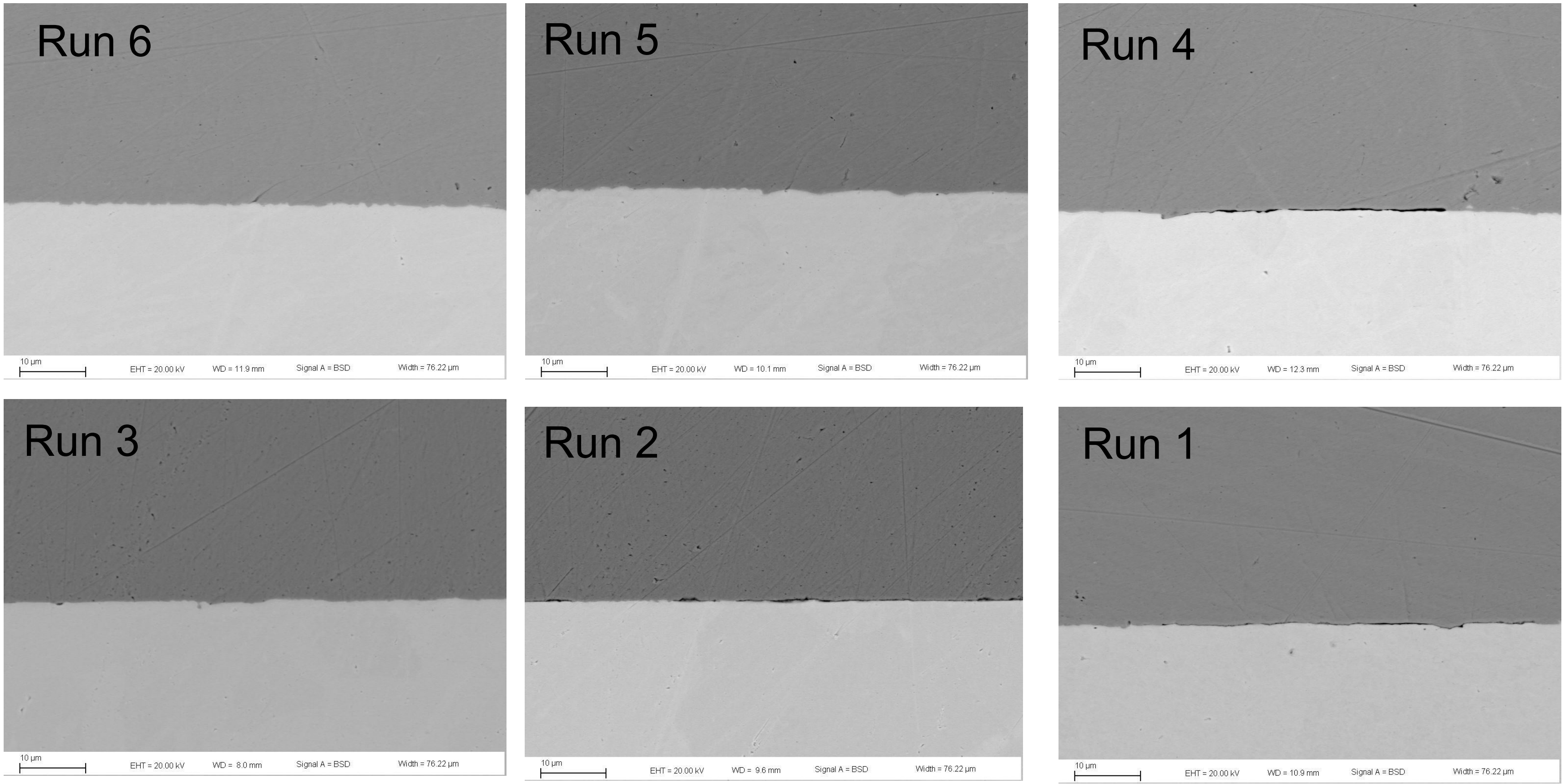


Run 1: 950ºC, 10.3 MPa, 0.5 hours



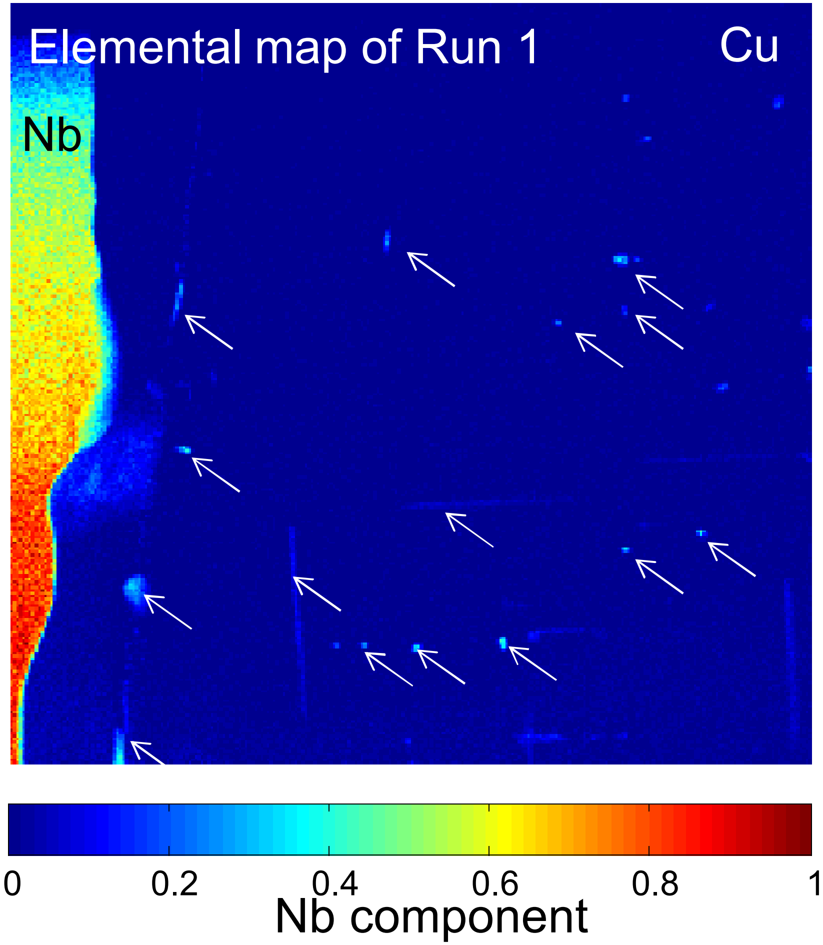
Run 2: 950ºC, 4.1 MPa, 3.0 hours

Interface comparison



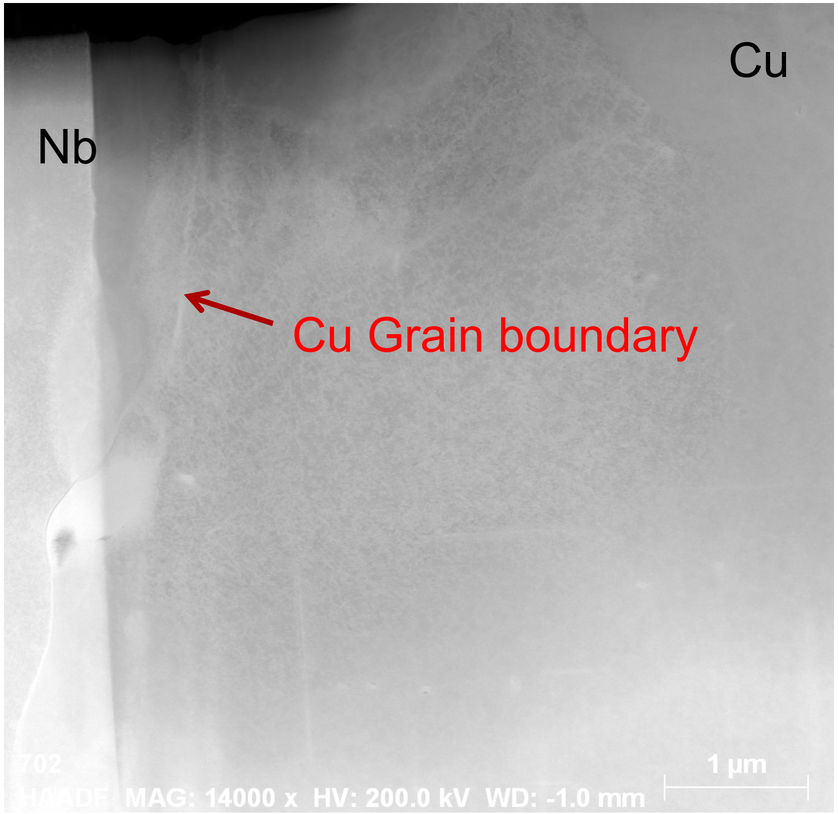
TEM analysis: Cu-Nb diffusion bond

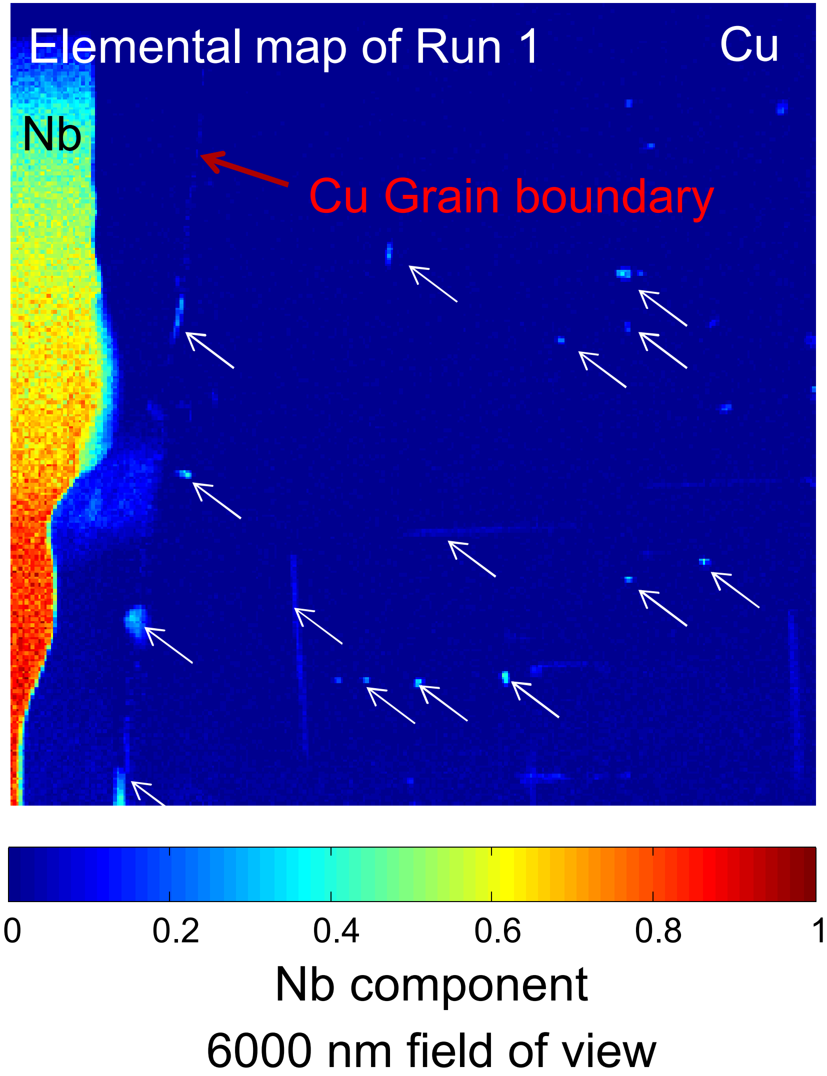
- TEM indicated Nb diffusion into the Cu and no Cu diffusion into the Nb.
  - Nb diffused along grain boundaries (high or low angle), and dislocations with in the Cu.
  - Nb diffusion was observed in all the samples subjected to TEM (Run 1, 3, 5, and 6)



6000 nm field of view

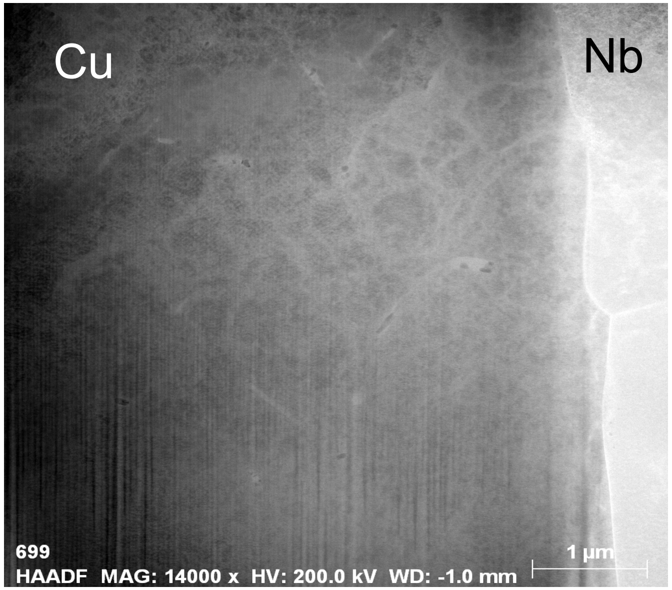
TEM analysis: Run 1(950ºC, 0.5 hrs)

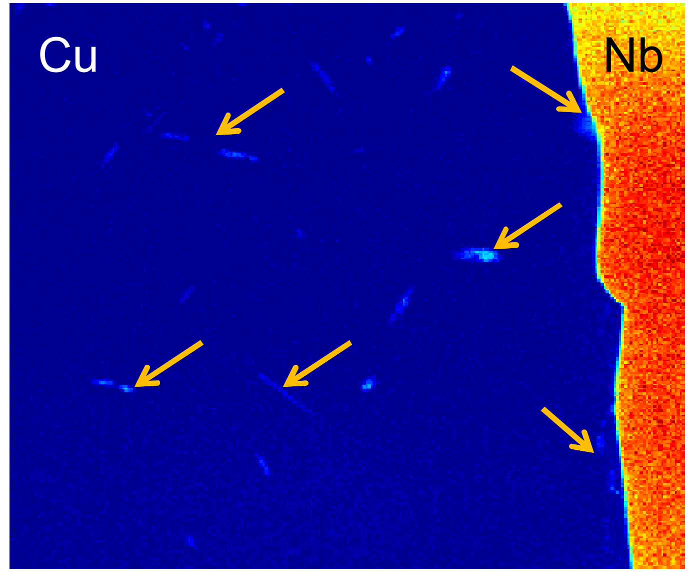


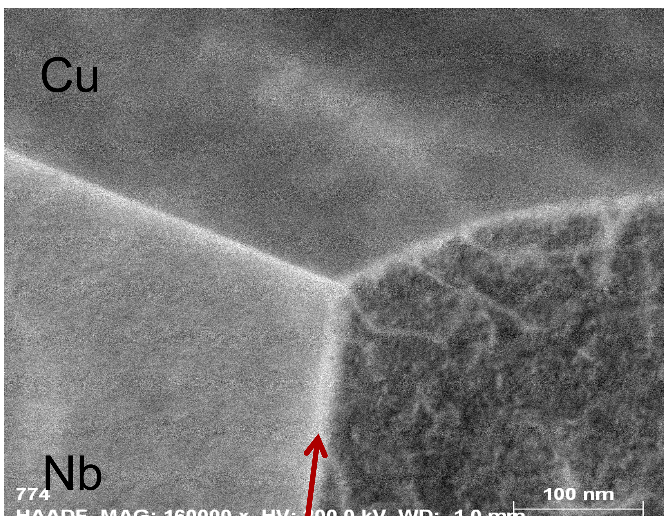


- Nb diffusion along grain boundary and dislocations.
- Nb diffusion 5-6µm from interface

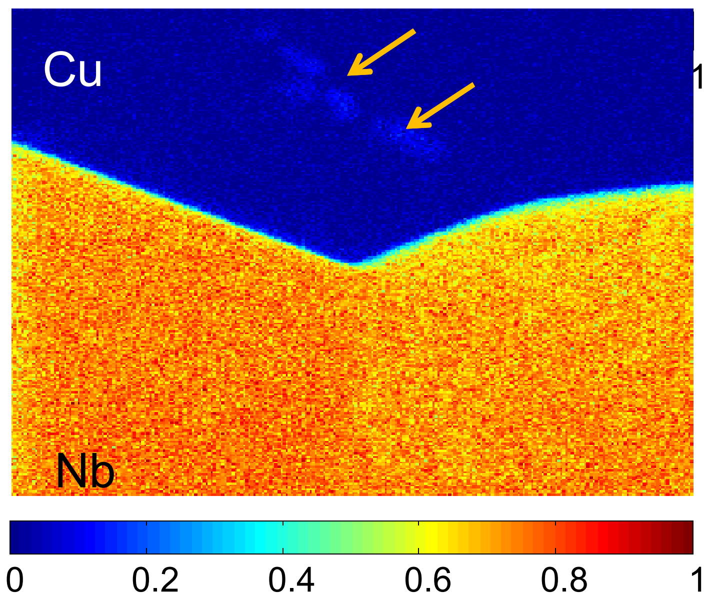
TEM analysis: Run 6 (1050^{o}C, 0.5 hrs)







Nb Grain boundary



Nb Component

6000 nm field of view

- Nb diffusion about 5µm from interface.
- No Cu along Nb grain boundary

Optimization experiments

- Experiments to determine optimal bonding parameters
  - Based of SEM results from initial experiments

	Temperature [°C]	Pressure [Mpa (KSI)]	Time [Hours]	Material thickness		Load applied [°C]
				Cu [μm]	Nb [μm]	
Run 7	1050	10.3 (1.5)	0.25	127	508	1050
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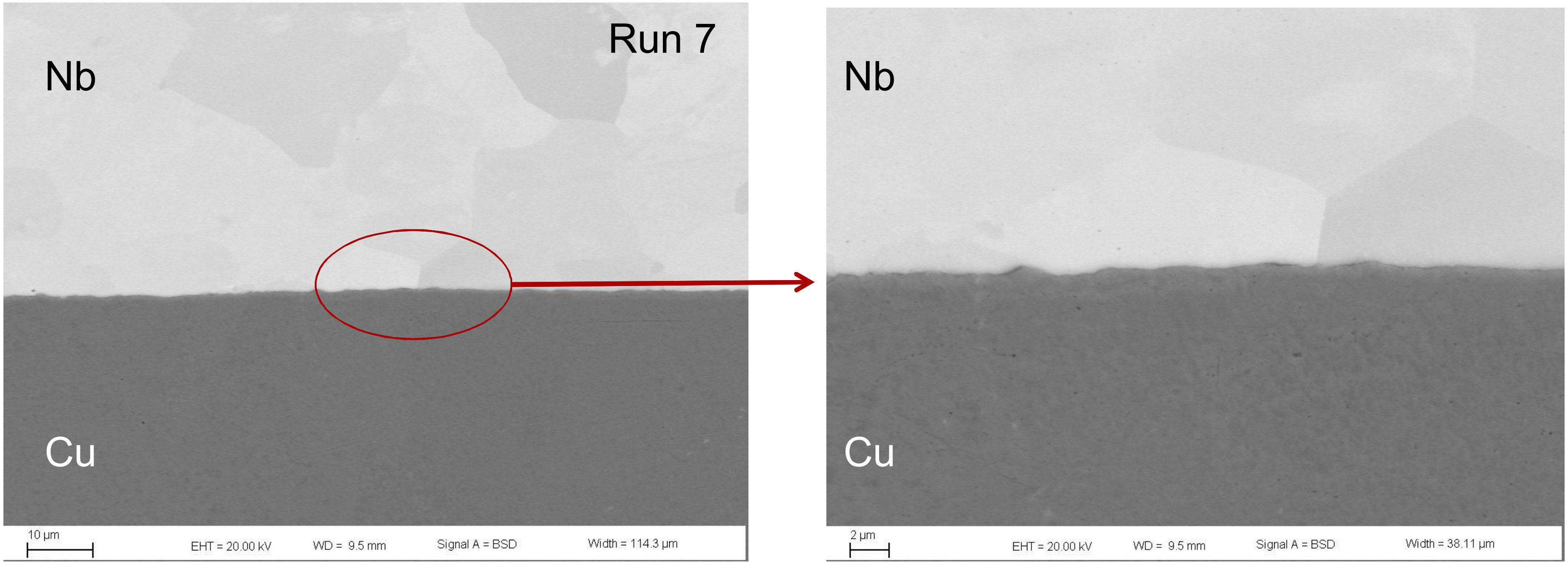
Run 9	1050	4.1 (0.6)	0.5	127	508	1050

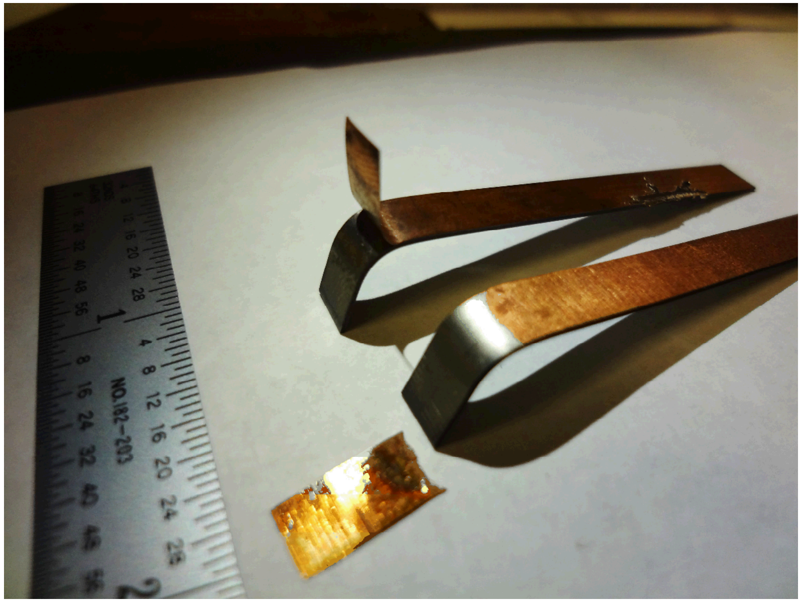
Optimization experiment results

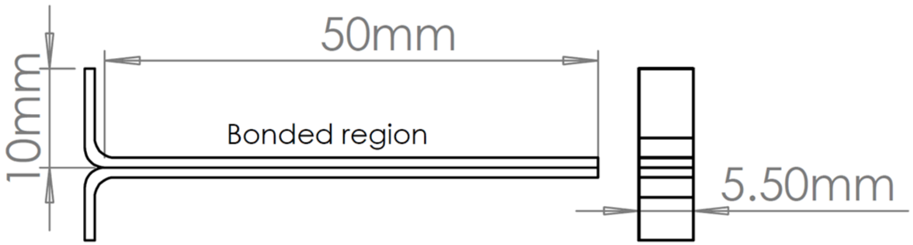


- Optimization experiment samples contained voids from ~1µm to ~40µm.
- Voids indicate the bonding time was insufficient.

Characterizing diffusion bond strength via peel testing

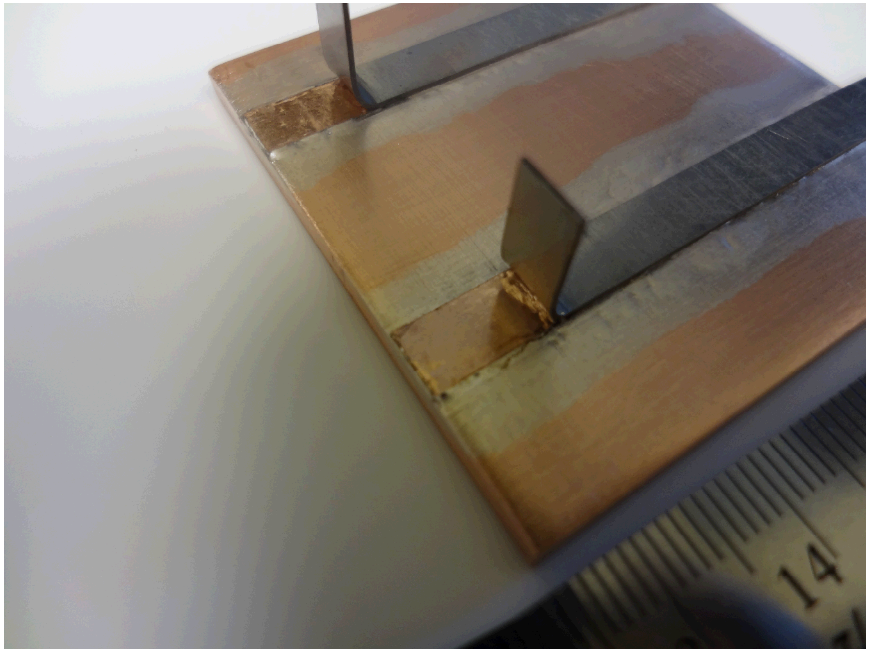
- Failure occurred in Cu between 18 and 20 pounds of force
  - All samples tested with the following orientation failed in the Cu at the Cu-Nb interface
  - The diffusion bond did not fail.

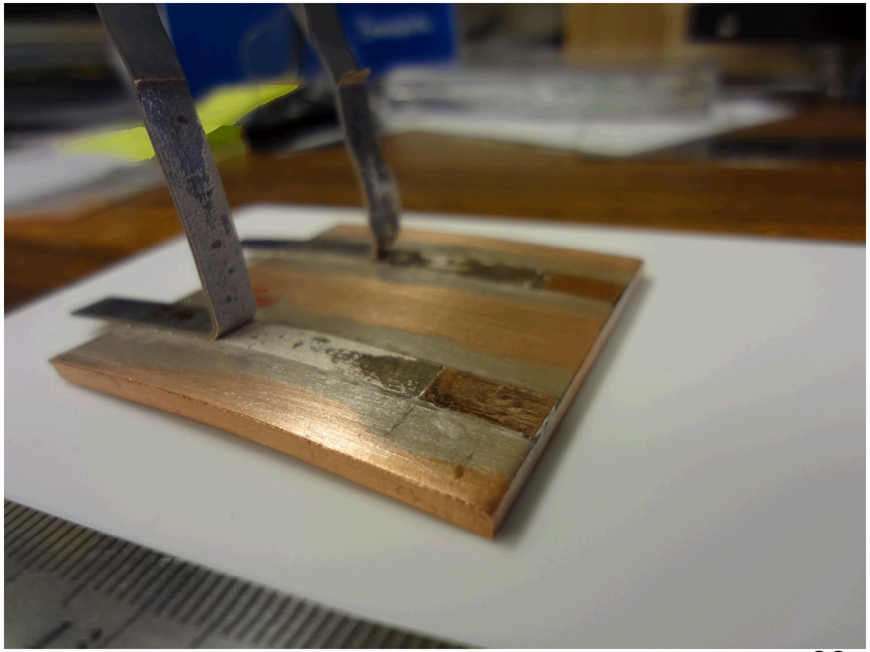




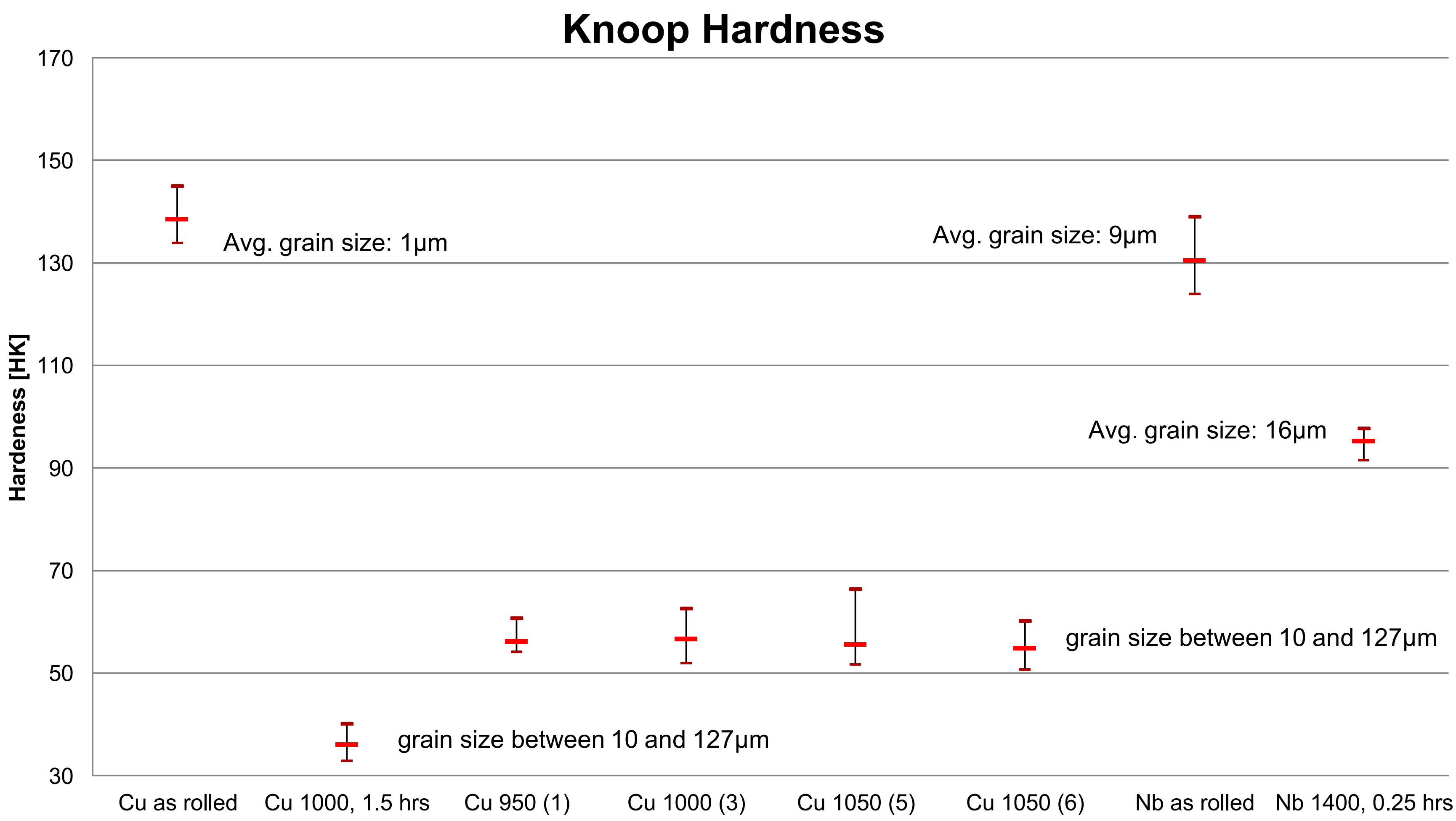
Peel test continued

- Additional attempts at testing the bond strength were inconclusive. Again, the diffusion bond did not fail.
- Cu-Nb strips were brazed to Cu using Cusil™ (brazing temp ~810^{o}C), failure of the braze joint occurred around 50 PSI.





Hardness test: Knoop Hardness



- 100gf used on all samples except ‘Cu 1000, 1.5 hrs’ where 50gf was used.

Experiments

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- Experiments conducted to determine best bonding parameters

	Temperature [°C]	Pressure [Mpa (KSI)]	Time [Hours]	Material thickness		Load applied [°C]
				Cu [μm]	Nb [μm]	
Run 1	950	10.3 (1.5)	0.5	127	508	950
Run 2	950	4.1 (0.6)	3	127	508	950
Run 3	1000	6.9 (1.0)	1.5	127	508	1000
Sample 1	1035	4.1 (0.6)	3	50.8	1016	1035
Run 4	1035	4.1 (0.6)	3	127	508	1035
Sample 2	1050	4.1 (0.6)	3	50.8	254	1050
Run 5	1050	4.1 (0.6)	3	127	508	1050
Run 6	1050	10.3 (1.5)	0.5	127	508	1050

Sample 1: SEM, and tensile buttons
Sample 2: SEM
Run 1,2,4: SEM
Run 3,5,6: SEM, and tensile buttons

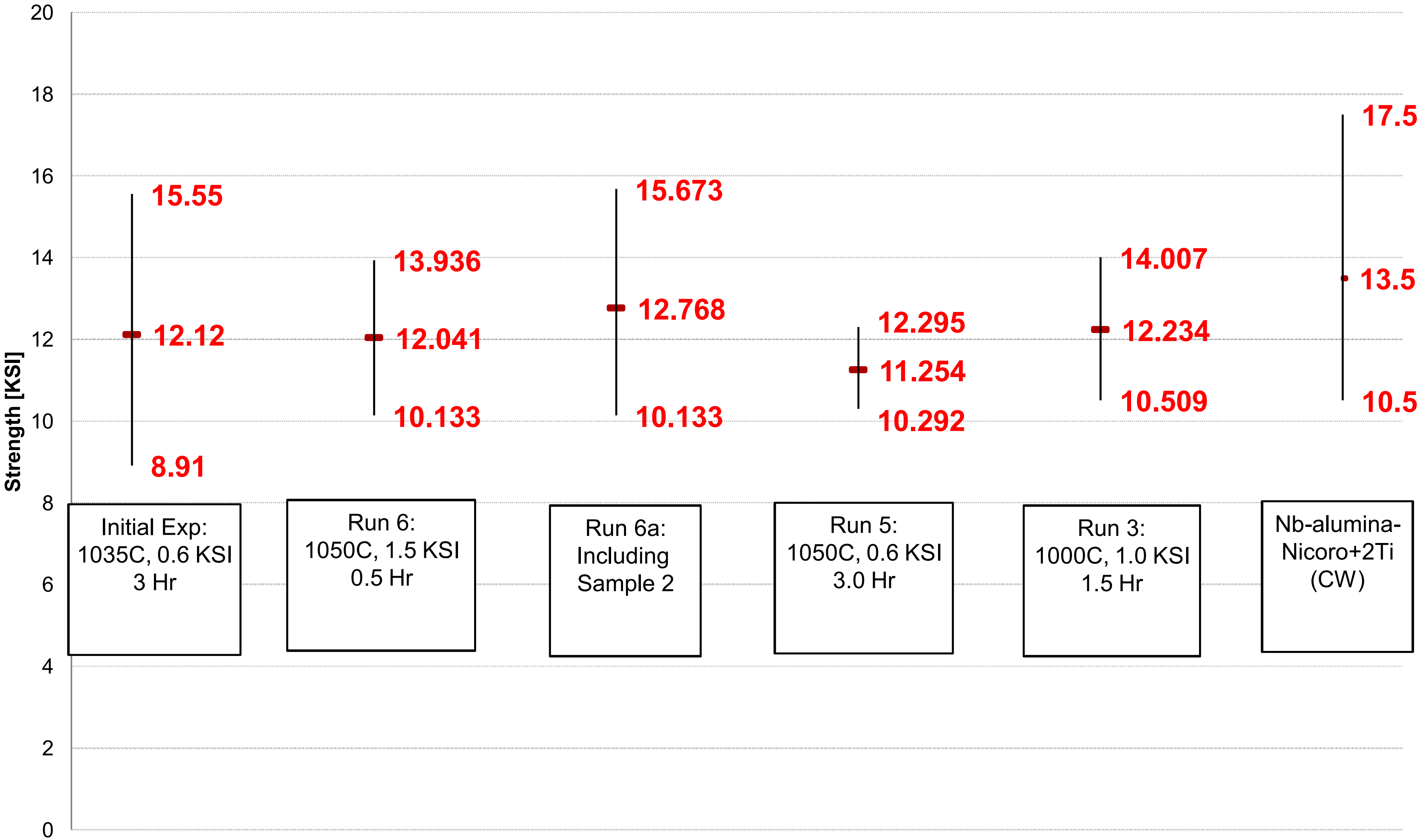
Tensile button test results

- Tensile button test
  - All tensile buttons were hermetic except one, which had a lopsided braze joint
  - Roughly 2/3 of samples failed on Cu side with crack initiation in ceramic, remaining 1/3 failed on the Nb side
  - Failure of tensile buttons initiated at or near braze fillets/blobs

	Avg. Load [lbs]	**Avg. KSI***	**Avg. MPa**
Run 6	2296.58	12.77	88.03
Run 5	2024.28	11.25	77.59
Run 3	2160.43	12.01	82.81
Initial (sample 1)	2180.50	12.12	83.58

*Surface area: 0.179873 sqr in

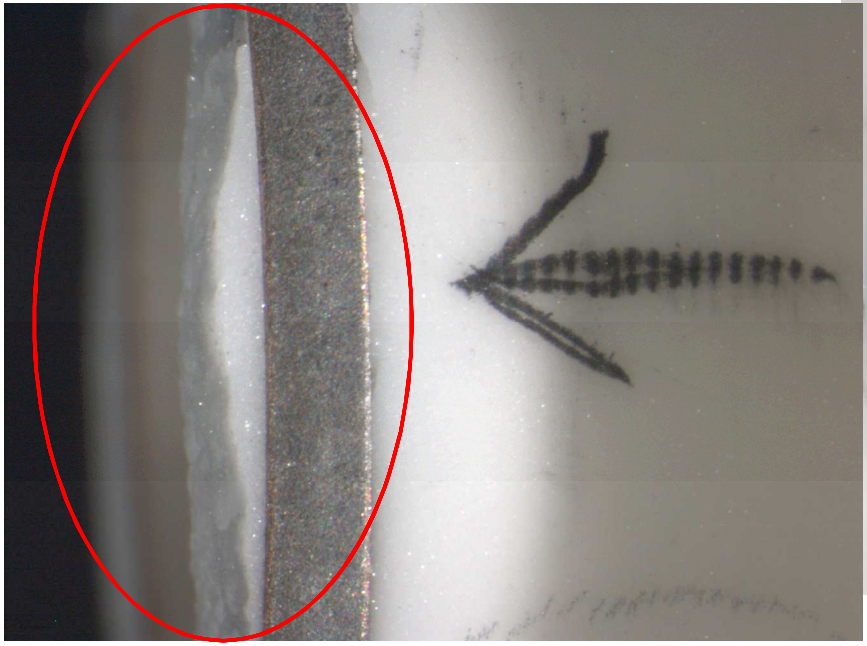
Tensile button test results

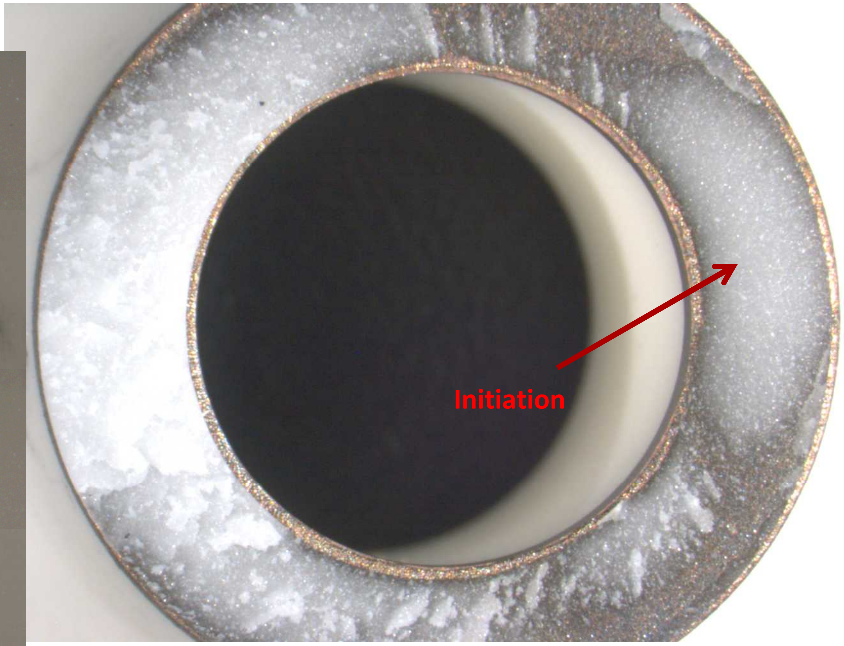


Results: Fracture of tensile button

- All tensile buttons had primary failure occur in the ceramic
- Sample tensile strength 107.21 MPa (15.5 KSI)

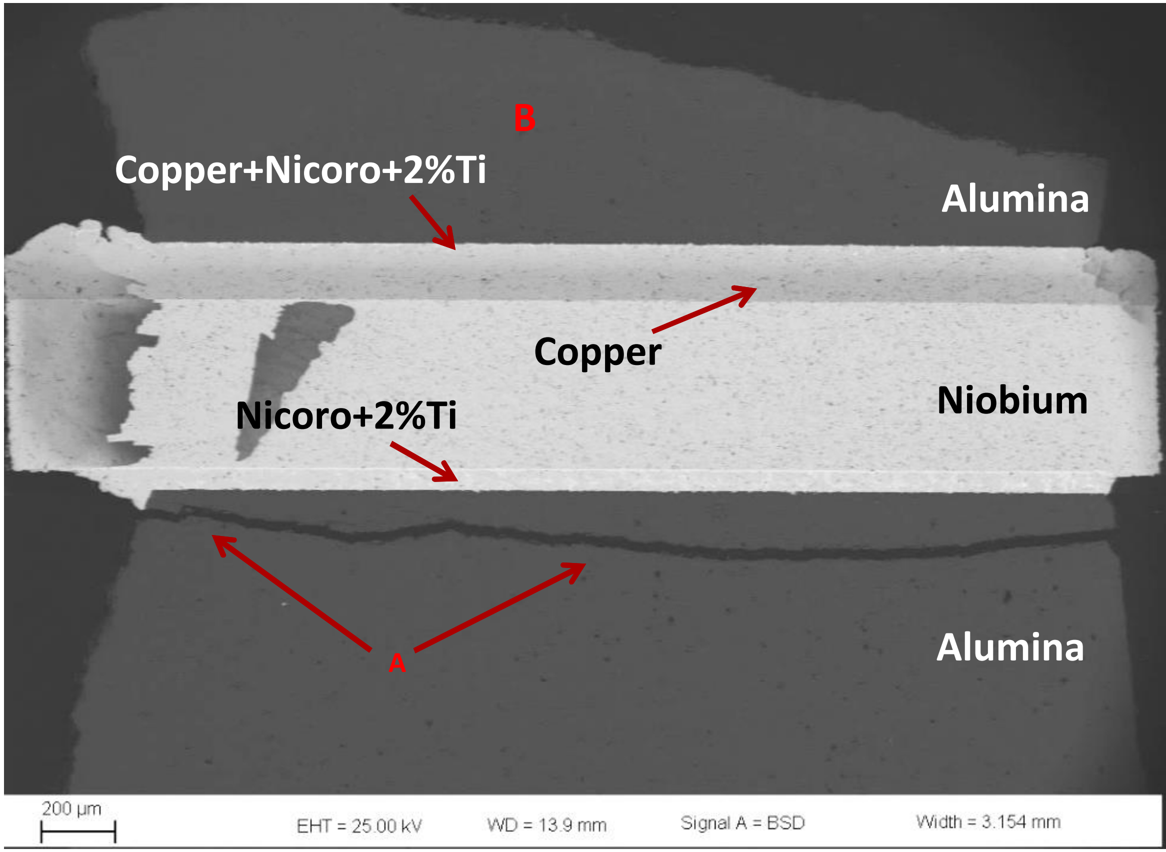
Sample 1-2





SEM of tensile button fracture

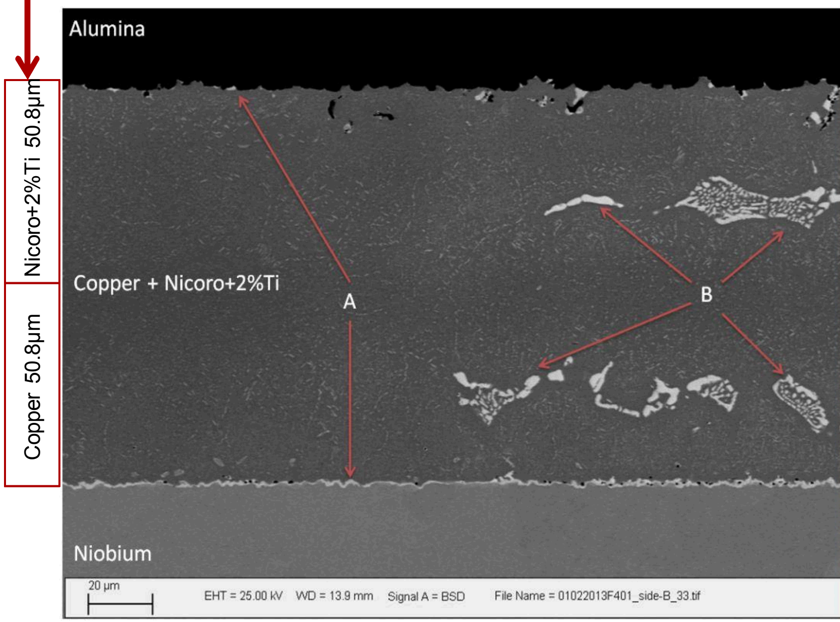
- 2/3 of the run 3, 5, and 6 samples tested had crack initiation (A) on the side opposite the primary failure (B).
- 75% of buttons with primary failure on Nb side exhibited cracks on Cu side.
- 64% of buttons with primary failure on Cu side exhibited cracks on Nb side.

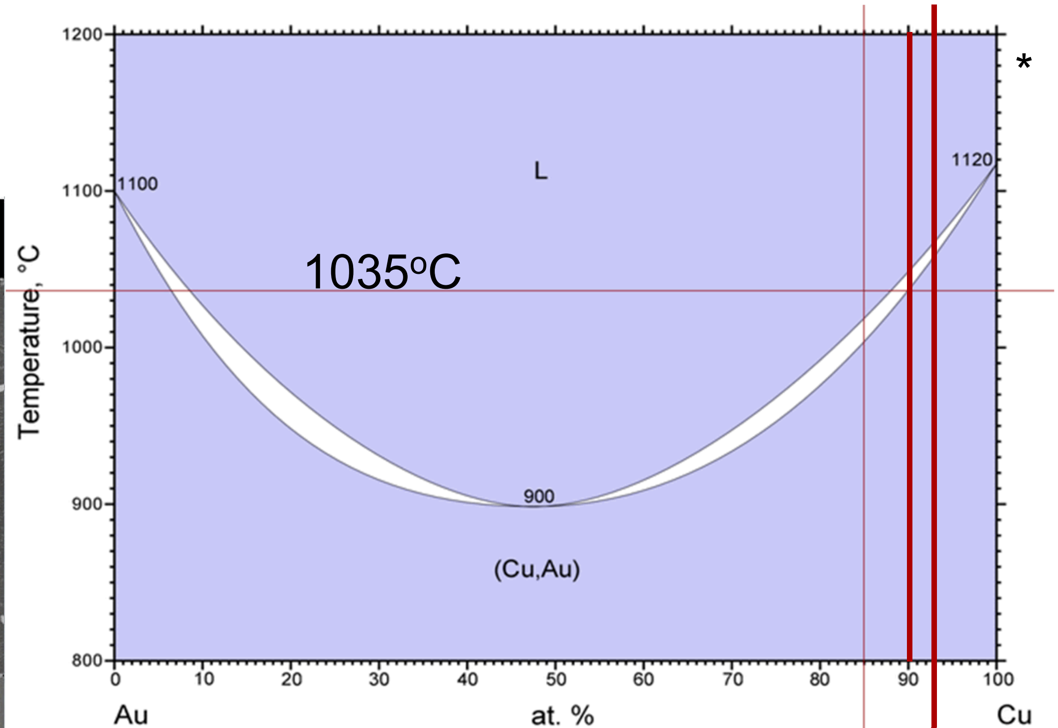


Results: Dissolution of Cu in braze joint

Original thickness

Cu-alumina braze joint after brazing





SEM of sample 1-2

Tensile strength: 107.21 Mpa

Original thicknesses:

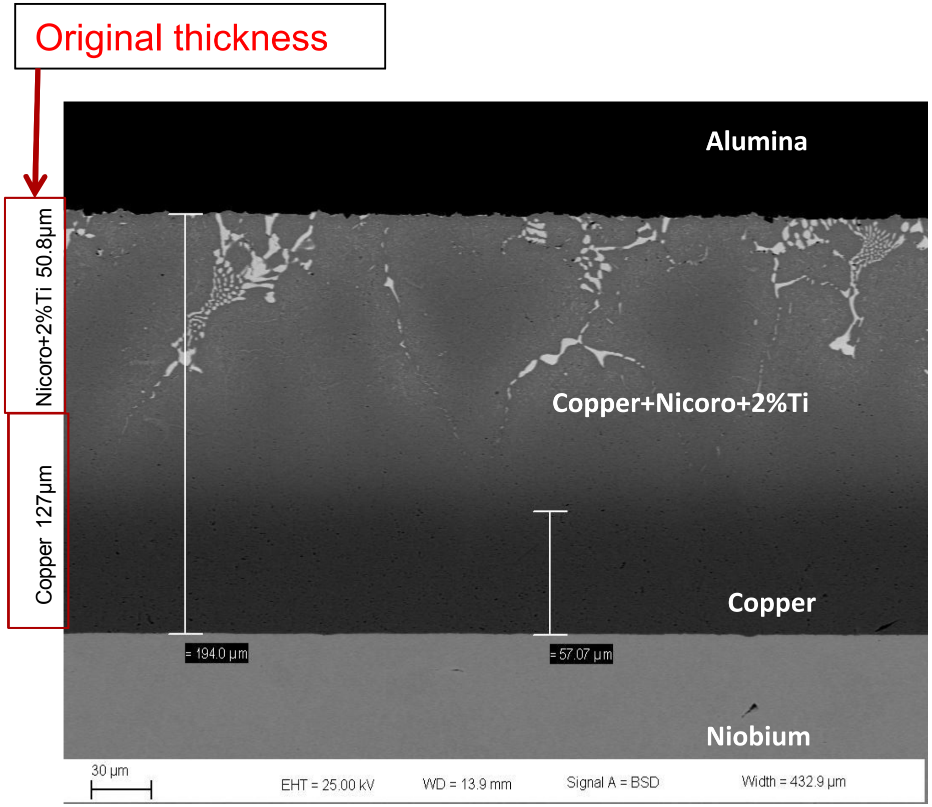
- Cu: 50.8µm, Nb: 1016µm
- Nicoro®+2%Ti: 50.8µm

Initial composition of braze alloy 85 at %Cu

Final composition of braze joint 93 at %Cu

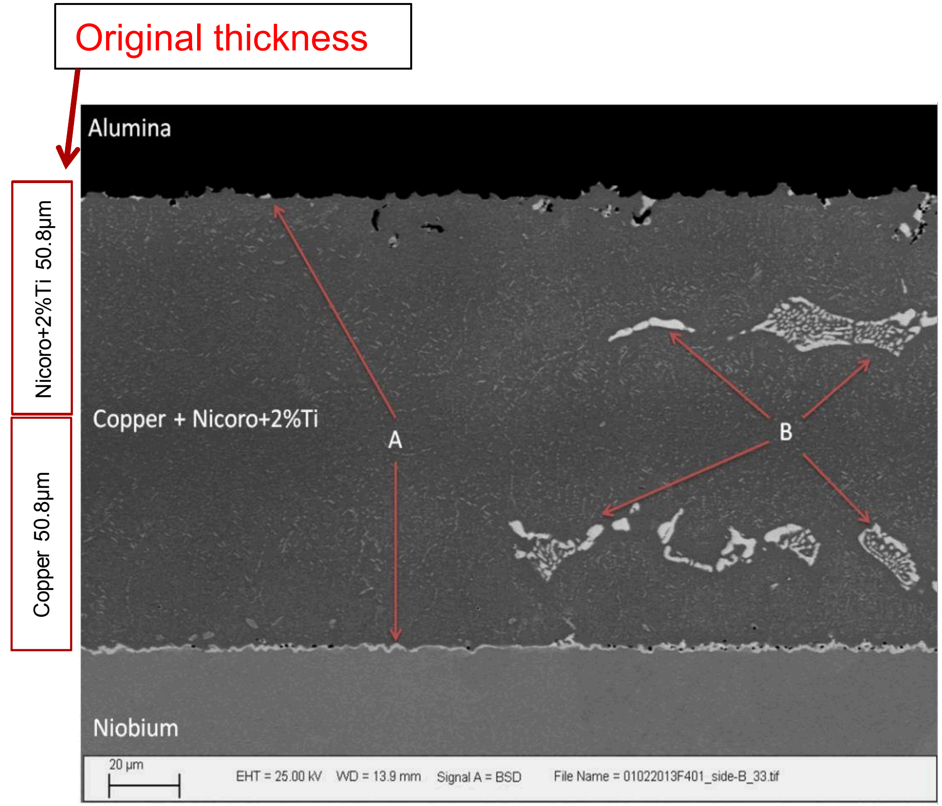
*W. T. Zheng, Study on phase diagrams for Ag-Au, Au-Cu binary alloys at high temperature, 36, 75-79, (1991)

Dissolution of Cu: 127µm Cu vs 50µm Cu



SEM of run 5- sample 2
Tensile strength: 76.9 Mpa
Failure occurred in ceramic
Original thicknesses:

Cu: 127 µm, Nb: 508 µm
Nicoro®+2%Ti: 50.8µm



SEM of sample 1-2
Tensile strength: 107.21 Mpa
Failure occurred in ceramic
Original thicknesses:

Cu: 50.8µm, Nb: 1016µm
Nicoro®+2%Ti: 50.8µm

Conclusions: Diffusion Bonding

- Diffusion bonding of Cu to Nb has been successfully demonstrated
  - Robust diffusion bonds of Cu to Nb can be formed under different conditions, given the conditions allow material creep to close all the voids
    - Optimal diffusion bonding parameters are: **1050ºC, 10.3 MPa, 0.5 hours**.
    - However, if the load required to achieve 10.3 MPa exceeds the limits of the hot press, then the following conditions can be used to achieve similar results: **1050ºC, 4.1 MPa, 3.0 hours**.
  - It has been demonstrated through tensile testing that the Cu-Nb diffusion bond is robust, withstanding tensile loads of at least 2200 lbs.

Conclusions: Brazing

- The complete dissolution is prevented when a thicker piece of Cu is used, intern increasing the amount of Cu in the braze joint.
- The complete dissolution of Cu in the braze joint and premature solidification at the braze temperature did not affect the braze strength.
- The partial dissolution of Cu in the braze joint did not cause the diffusion bonded interface to fail during tensile testing.

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Questions and Comments