JOINING AND INTEGRATION OF SILICON NITRIDE CERAMICS FOR AEROSPACE AND ENERGY SYSTEMS

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Abstract

Light-weight, creep-resistant silicon nitride ceramics possess excellent high-temperature strength and are projected to significantly raise engine efficiency and performance when used as turbine components in the next-generation turbo-shaft engines without the extensive cooling that is needed for metallic parts. One key aspect of Si_3N_4 utilization in such applications is its joining response to diverse materials. In an ongoing research program, the joining and integration of Si_3N_4 ceramics with metallic, ceramic, and composite materials using braze interlayers with the liquidus temperature in the range 750-1240C is being explored. In this paper, the self-joining behavior of Kyocera Si_3N_4 and St. Gobain Si_3N_4 using a ductile Cu-based active braze (Cu-ABA) containing Ti will be presented. Joint microstructure, composition, hardness, and strength as revealed by optical microscopy, scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), Knoop microhardness test, and offset compression shear test will be presented. Additionally, microstructure, composition, and joint strength of Si_3N_4/Inconel 625 joints made using Cu-ABA, will be presented. The results will be discussed with reference to the role of chemical reactions, wetting behavior, and residual stresses in joints.

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Outline

- **Introduction and Background**
- **Technical Challenges**
  - ***Wetting and Reactions***
  - ***Thermal Expansion Mismatch***
- **Experimental Procedure**
  - ***Active Metal Brazing***
  - ***Characterization (SEM, EDS)***
  - ***Mechanical Testing***
- **Results and Discussion**
  - ***Joint Microstructure***
  - ***Mechanical Behavior***
- **Concluding Remarks**

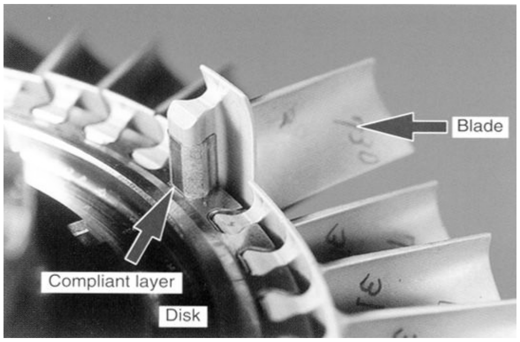
Need for Joining and Integration of Silicon Nitride Ceramics to Itself and to Metallic Systems

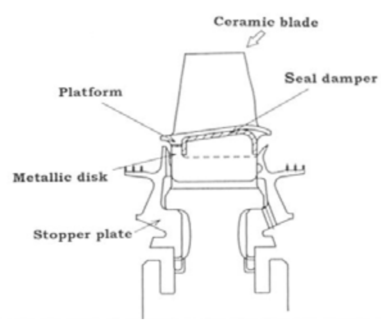
- Joining and integration is an enabling technology for the manufacturing and application of advanced ceramic components in aerospace and energy systems.
- Robust joining technologies for Silicon Nitride to itself, using high temperature (>1300°C) capable ceramic interlayers, could play a key role in low cost manufacturing of complex shaped components.
- Bonding of Silicon Nitride to metals (stainless steels, Fe alloys, Mo, Nickel, etc.) has been carried out extensively over the last few decades. However, poor wettability of ceramics (poor flow and spreading characteristics) and thermoelastic incompatibilty always provide significant challenges.
- Integration of Silicon Nitride to metals in components and systems requires the development and validation of innovative joining concepts and technologies, which are capable of higher operating temperatures.

Technical Challenges in Integration of Ceramic-Metal *vs* Ceramic-Ceramic Systems

Ceramic-Metal System		Ceramic-Ceramic System
• Flow and wettability		• Reaction and diffusion
• Roughness	Common Issues	• Roughness
• Residual stress (ΔCTE)		• Residual stress (ΔCTE)
• Multi-axial stress state		• Multi-axial stress state
• Joint design		• Joint design
• Joint stability in service		• Joint stability in service
• Metal – *forgiving*		• Ceramic – *unforgiving*
• Elastic-plastic system		• Elastic-elastic system
• Lower use temperatures		• Higher use temperatures
• Less aggressive environment		• More aggressive environment

Silicon Nitride Based Components for Energy Systems





Hybrid Gas Turbine Blade (Ceramic Blade and Metallic Disk)
in NEDO's Ceramic Gas Turbine R&D Program, Japan (1988-1999)

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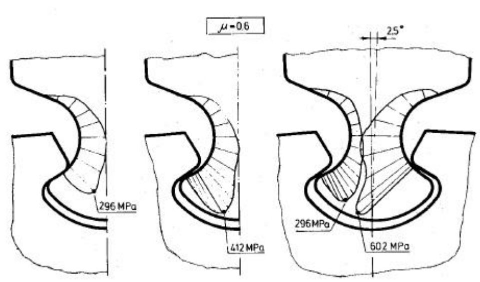
Integration Technologies for Silicon Nitride Ceramics to Metallic Components

Issues with Ceramic Inserted Blades



There are contact stresses at the metal-ceramic interface.
Compliant layers (i.e. Ni-alloy+Pt) are used to mitigate the
stress and damage. Failures can occur in the compliant layer.

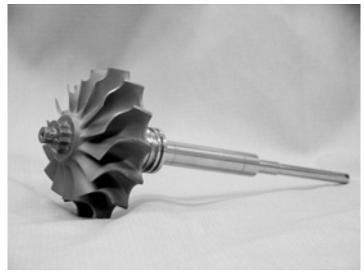
Mark van Roode, "Advances in the Development of Silicon Nitride and Other Materials", Environmental Barrier Coatings Workshop, November 6, 2002, Nashville, TN.

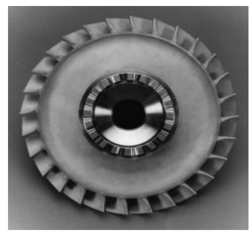
Integration Technologies for Silicon Nitride Ceramics to Metallic Components

INTEGRAL ROTORS

- No Compliant Layer with Disk
- Attachment of Ceramic Rotor to Metal Shaft
- Primarily Small Parts
- Ability to Fabricate Larger Parts Has Improved
- Integral Rotors are Replacing Metal Disks with Inserted Blades

Industry Direction





IR Silicon Nitride Rotor, DOE Microturbine Program (top) H-T. Lin, ORNL

Mark van Roode, Solar Turbines

Ongoing Activities in Integration of Silicon Nitride Ceramics to Metals Using Metallic Interlayers

Active Metal Brazing

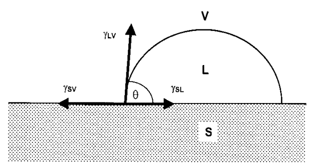
Metallic Systems:
- Inconel-625 and other Ni-Base Superalloys
- Cu-clad Mo, Ti

Si_3N_4- Based Ceramics:
- Saint Gobain NT-154
- Kyocera SN 281, 282
- Sialons

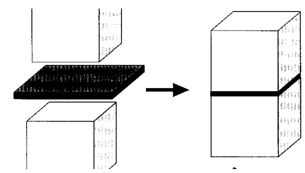
Interlayer Systems:
- Active Metal Brazes (Ag, Cu, and Pd based)
- Metallic Glass Ribbons

Technical Issues:
- **Melting range / behavior**
- **Wetting characteristics**
- **Atmosphere compatibility**
- **Compositional compatibility**
- **Cost & availability**

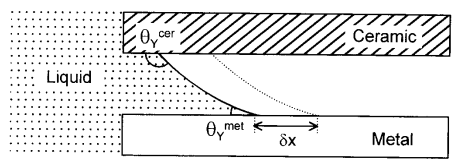
Wettability is Important Factor in Brazing



Contact angle of braze should be small



Braze layer melts and spreads between the substrates to form the joint



Ordinary braze alloys wet the metal but not the ceramic!

Must use 'active' brazes that wet and bond with both metal and ceramics

Objective

- **Utilize brazing approach to bond silicon nitride ceramics to itself and to metals using active braze alloys.**
- **Characterize the joint microstructure and composition at the joint interface.**
- **Evaluate the mechanical behavior of joints at different temperatures.**

Experimental Procedure - Materials -

- **Silicon Nitride Ceramics:**
  - *Kyocera SN-281: contains ~ 9-10% wt% Lu_2O_3*
  - *Saint Gobain NT 154: contains ~ 4 wt% Y_2O_3*
- **Inconel 625**
  - *Inco Specialty Metals*
  - *Nominal composition (in wt%): 58Ni-21.5Cr-9Mo-5Fe-1Co-0.5Si-0.5Mn-0.4Al-0.4Ti*
- **Braze alloy: Cu-ABA**
  - *Morgan Advanced Ceramics, Hayward, CA.*
  - *Braze foil thickness ~ 50 μm*

Braze Composition, (wt %)	T_L, K	T_S, K	E, GPa	YS, MPa	UTS, MPa	CTE, ×10^{-6} K^{-1}	% El.
Cu-ABA® (92.75Cu-3Si-2Al-2.25Ti)	1297	1231	96	278	620	19.6	42

- **Intermediate Layers**
  - *Thin (5 μm) intermediate layers of pure (99.97%) Cu in some joints*

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Experimental Procedure

- **Substrates and braze foils cut into 2.54 cm x 1.25 cm x 0.25 cm panels and ultrasonically cleaned.**
- **Two braze foils sandwiched between substrates and heated under vacuum (~10^{-6} torr) to 15-20°C above T_L. After soak (5 min. or 30 min.), joints were slowly cooled.**
- **Brazed joints mounted in epoxy, ground, polished, and examined using optical microscopy and Scanning Electron Microscopy (JEOL JSM-740A) coupled with EDS.**
- **Shear strength test done on offset joints under compressive loading on an Instron 8562 machine using hydraulic grip platens, a SS316 die, and a deflectometer (Instron LVDT 2602-061). Loading rate: 50N/s.**
- ***Only Saint Gobain (NT-154) joints were tested because of its commercial availability.***

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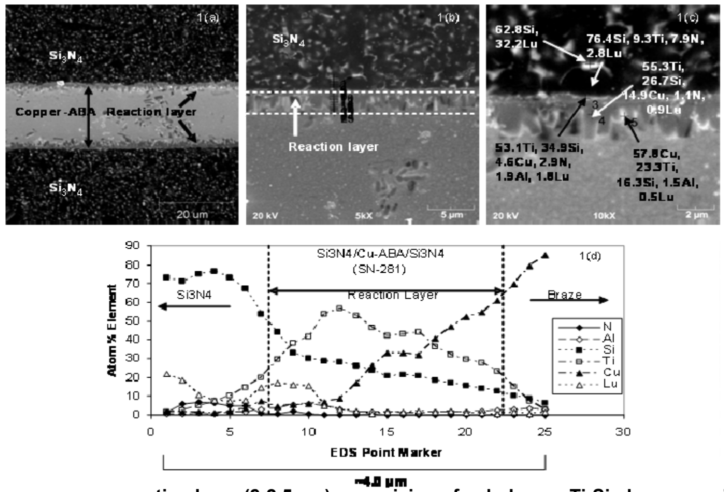
Self-joined Kyocera SN-281 Si_3N_4 brazed at 1317 K for 5 min

EDS data in (d) correspond to the point markers in (b)



- An inhomogeneous reaction layer (2-2.5 µm) comprising of a dark-gray Ti-Si phase, possibly titanium silicide, and a lighter Cu (Si, Ti) phase has developed.
- The product phase crystals are oriented perpendicular to the interface (growth direction).
- No interfacial excess of Lu; exists in minute quantities in reaction layer and increases toward Si_3N_4 .

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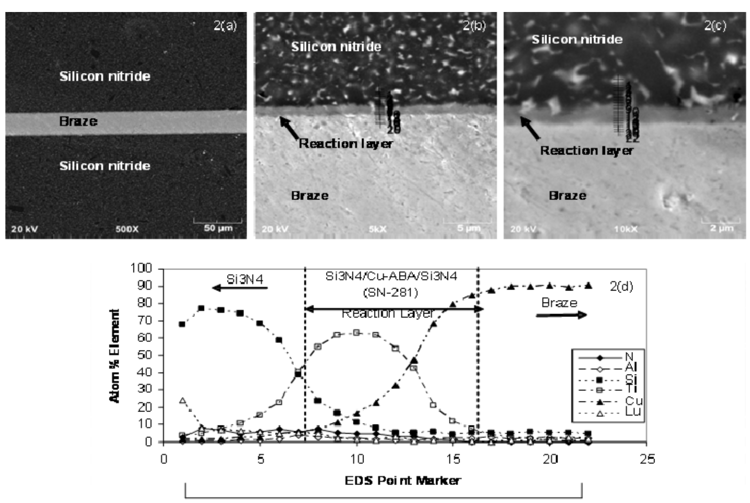
Self-joined Kyocera SN-281 Si_3N_4 brazed at 1317 K for 30 min

EDS data in (d) correspond to the point markers in (C)



- No increase in reaction layer thickness for 30 min. (faster kinetics in the early stages of reaction).
- Morphologically a more homogeneous, compact, and featureless reaction layer (possible coalescence of coarsened silicide crystals).

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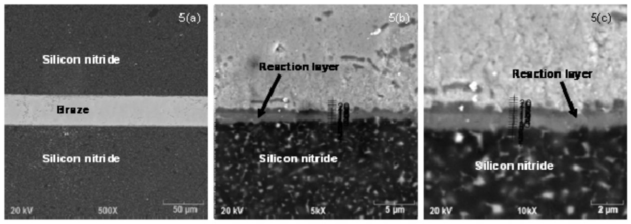
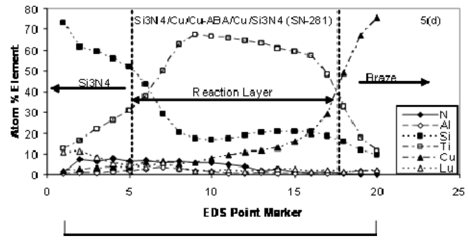
Self-joined Kyocera SN-281 brazed at 1317K for 30 min. (with 5 µm thick Cu foil inserts: $Si_3N_4/Cu/Cu\text{-}ABA/Cu/Si_3N_4$)

EDS data in (d) correspond to the point markers in (c)





- **Sound joint with a compact and morphologically homogeneous reaction layer (~1-2 µm thick).**
- **Ti and Si enrichments at the interface (possible formation of a titanium silicide compound layer).**

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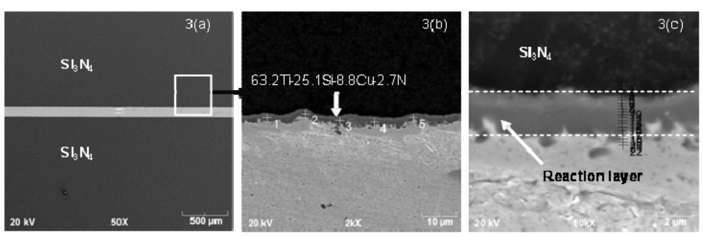
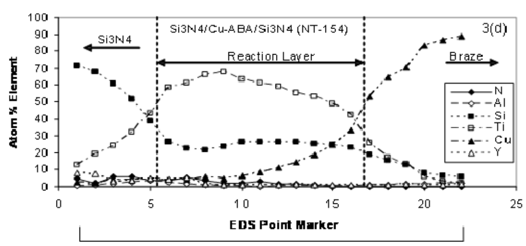
Self-joined St. Gobain NT-154 Si_3N_4 brazed at 1317 K for 30 min

EDS data in (d) correspond to the point markers in (c)





- **30 min. brazing time led to a homogenous, featureless reaction layer (slightly thicker, 2.5-3.5 µm) than the layer in the Kyocera joints.**
- **Reaction layer is rich in Ti and Si (approx. atomic concentration of the layer is 63Ti-25Si-9Cu-3N).**

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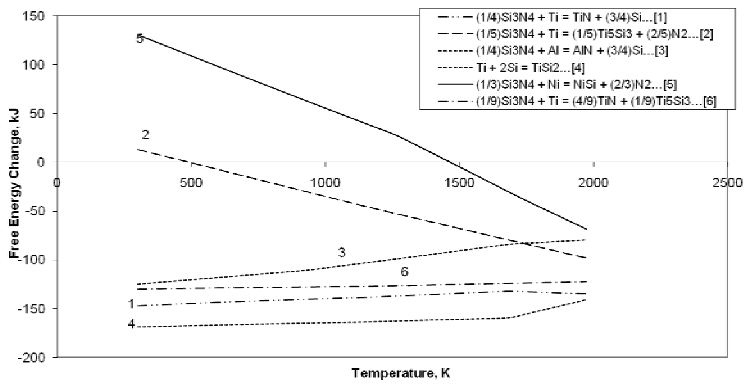
Gibb's Free Energy Change for Reactions of Si_3N_4 with Ti, Al, Ni and Si

Calculations using the software HSC Chemistry version 4.1 (Outokumpu Ra, Oy, Finland)



- Titanium nitride and titanium silicides could form in the joint region.

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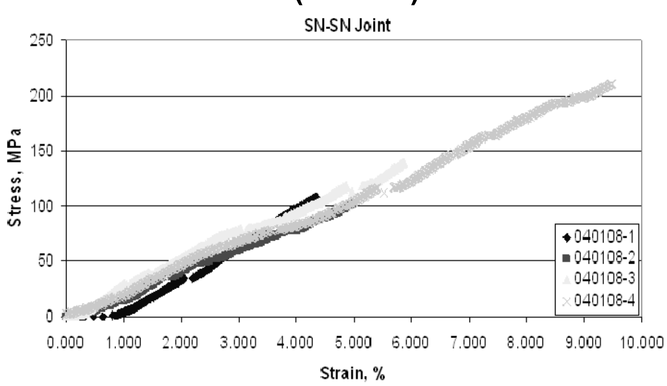
Stress-Strain Behavior of Si_3N_4/Cu-ABA/Si_3N_4 Joints (NT-154)



- Linear stress-strain behavior.
- Fracture stress is in the range 103-211 MPa.
- Mean fracture stress is 140.5 MPa (standard deviation: 49.6 MPa).
- Fracture propagated through Si_3N_4 (not through the joint region).

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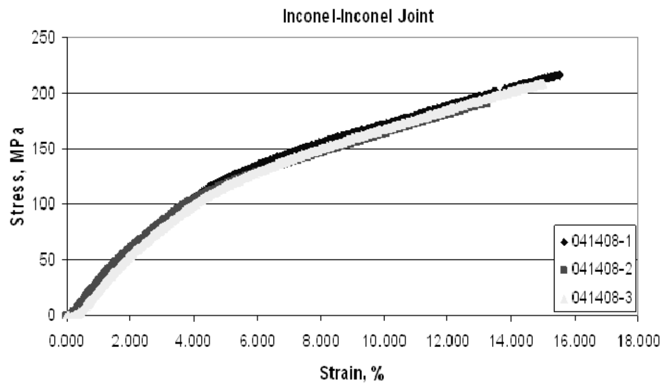
Stress-Strain Behavior of Inconel/Cu-ABA/Inconel Joints



- Fracture stress is in the range 191-221 MPa.
- Mean fracture stress is 206.7 MPa (standard deviation: 12.3 MPa).
- Joints failed through the braze region at the bonded interface.

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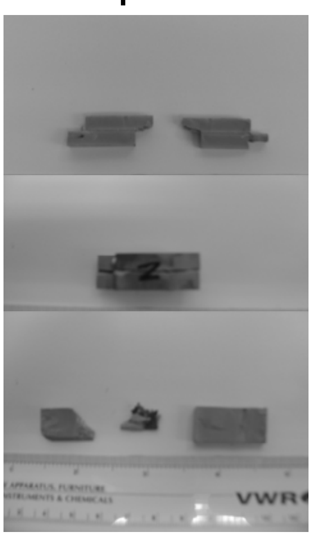
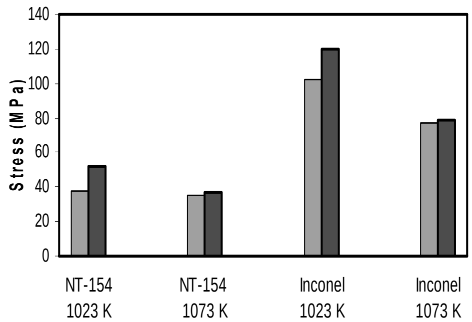
Post Test Photographs of Si_3N_4/Cu-ABA/Si_3N_4 Specimens Tested at Room Temperature

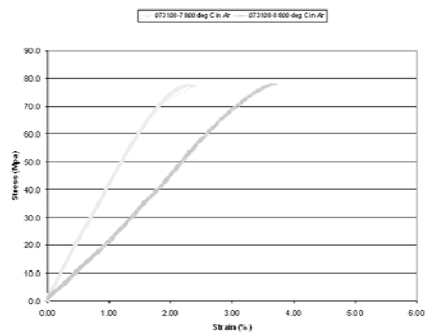


RT Tested Specimens

High Temperature Test Set-up

Strength of Si_3N_4/Cu-ABA/Si_3N_4 (NT 154) and Inconel-Inconel Joints at 1023 and 1073 K





- The strength of silicon nitride joints drops to around 35 MPa at 1073 K due to thermal expansion mismatch and potential degradation in braze properties.
- Strength reduction is similar in Inconel-Inconel joints although the strength values are higher using the same braze (~78 MPa).

Observations on the Mechanical Behavior of Silicon Nitride/Cu-ABA/Inconel 625 Joints

- **Directly bonded silicon nitride (NT-154)/Cu-ABA/Inconel 625 specimens exhibit lower strengths (~15 MPa) due to thermal stresses generated by widely varying thermal expansion coefficients of constituents.**
- **Ductile metallic multilayers will be needed to accommodate the residual stresses due to mismatch in thermal expansion coefficients.**
- **Activities are underway to utilize metallic multilayers with different thermal expansion coefficients and yield strengths to control the residual stresses.**
- **Preliminary mechanical test results are very promising and will be reported at a later date.**

Concluding Remarks

- Self-joining of Kyocera Si_3N_4 (SN-281) and St. Gobain Si_3N_4 (NT-154) using a ductile Cu-Al-Si-Ti active braze was demonstrated.
- Under identical joining conditions (1317 K, 30 min), the Si_3N_4/braze reaction layer was slightly thicker (~2.5-3.5 µm) in NT-154 joints than in SN-281 joints (~1.0-2.0 µm).
- There was no interfacial excess of either Lu (in Kyocera Si_3N_4) or Y (in St. Gobain Si_3N_4) at the Si_3N_4/braze interface. The interfaces were enriched in Ti and Si possibly due to the formation of titanium silicides.
- Thin (~5.0 µm) Cu foils placed at the Si_3N_4/braze interface did not alter either the thickness of the reaction layer or its composition.
- The morphological homogeneity of the interfacial layers increased with increasing brazing time, and led to a featureless reaction zone in 30 min. due possibly to coalescence of the product phase crystals.
- Continuing research is focusing on joining and characterization of Si_3N_4 /Inconel joints using multilayer bonding approaches.