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$_3$N$_4$ utilization in such applications is its joining response to diverse materials. In an ongoing research program, the joining and integration of Si$_3$N$_4$ ceramics with metallic, ceramic, and composite materials using braze interlayers with the liquidus temperature in the range 750-1240°C is being explored. In this paper, the self-joining behavior of Kyocera Si$_3$N$_4$ and St. Gobain Si$_3$N$_4$ 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$_3$N$_4$/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.
Joining and Integration of Silicon Nitride Ceramics for Aerospace and Energy Systems

M. Singh* and R. Asthana**

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

** Department of Engineering & Technology
University of Wisconsin-Stout
Menomonie, WI 54751

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 incompatibility 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

<table>
<thead>
<tr>
<th>Ceramic-Metal System</th>
<th>Ceramic-Ceramic System</th>
<th>Common Issues</th>
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<tbody>
<tr>
<td>- Flow and wettability</td>
<td>- Reaction and diffusion</td>
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<tr>
<td>- Roughness</td>
<td>- Roughness</td>
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<td>- Residual stress (ΔCTE)</td>
<td>- Residual stress (ΔCTE)</td>
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<td>- Multi-axial stress state</td>
<td>- Multi-axial stress state</td>
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<tr>
<td>- Joint design</td>
<td>- Joint design</td>
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<tr>
<td>- Joint stability in service</td>
<td>- Joint stability in service</td>
<td></td>
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<tr>
<td>- Metal – forgiving</td>
<td>- Ceramic – unforgiving</td>
<td></td>
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<tr>
<td>- Elastic-plastic system</td>
<td>- Elastic-plastic system</td>
<td></td>
</tr>
<tr>
<td>- Lower use temperatures</td>
<td>- Higher use temperatures</td>
<td></td>
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<tr>
<td>- Less aggressive environment</td>
<td>- More aggressive environment</td>
<td></td>
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</tbody>
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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)

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**

Mark van Roode, Solar Turbines

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

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

**Si₃N₄-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

IR Silicon Nitride Rotor, DOE Microturbine Program (top) H-T. Lin, ORNL
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% Lu₂O₃
  - Saint Gobain NT 154: contains ~ 4 wt% Y₂O₃

- Inconel 625
  - Newco 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

<table>
<thead>
<tr>
<th>Braze Composition (wt%)</th>
<th>Tₜ K</th>
<th>Tₜp K</th>
<th>E₀ GPa</th>
<th>Y₀ MPa</th>
<th>UTS, MPa</th>
<th>CTE, x10⁴ K⁻¹</th>
<th>% EL</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu-ABA (62.75Cu-36.8Al-0.35Ni)</td>
<td>1237</td>
<td>1231</td>
<td>96</td>
<td>270</td>
<td>630</td>
<td>19.6</td>
<td>42</td>
</tr>
</tbody>
</table>

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

• 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 Tₜ. 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.
• 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 Si₃N₄.

Self-joined Kyocera SN-281 Si₃N₄ brazed at 1317 K for 5 min
EDS data in (d) correspond to the point markers in (b)

• 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).

Self-joined Kyocera SN-281 Si₃N₄ brazed at 1317 K for 30 min
EDS data in (d) correspond to the point markers in (C)
Self-joined Kyocera SN-281 brazed at 1317K for 30 min.
(with 5 µm thick Cu foil inserts: Si$_3$N$_4$/Cu/Cu-ABA/Cu/Si$_3$N$_4$)

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).

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

- 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 Si₃N₄ (not through the joint region).
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.

Post Test Photographs of Si$_3$N$_4$/Cu-ABA/Si$_3$N$_4$
Specimens Tested at Room Temperature

RT Tested Specimens

High Temperature Test Set-up
Strength of Si₃N₄/Cu-ABA/Si₃N₄ (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 Si₃N₄ (SN-281) and St. Gobain Si₃N₄ (NT-154) using a ductile Cu-Al-Si-Ti active braze was demonstrated.

- Under identical joining conditions (1317 K, 30 min), the Si₃N₄/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 Si₃N₄) or Y (in St. Gobain Si₃N₄) at the Si₃N₄/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 Si₃N₄/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 Si₃N₄/Inconel joints using multilayer bonding approaches.