Ceramic Integration Technologies for Aerospace and Energy Systems: Technical Challenges and Opportunities

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Abstract

Ceramic integration technology has been recognized as an enabling technology for the implementation of advanced ceramic systems in a number of high-temperature applications in aerospace, power generation, nuclear, chemical, and electronic industries. Various ceramic integration technologies (joining, brazing, attachments, repair, etc.) play a role in fabrication and manufacturing of large and complex shaped parts of various functionalities. However, the development of robust and reliable integrated systems with optimum performance requires the understanding of many thermochemical and thermomechanical factors, particularly for high temperature applications. In this presentation, various challenges and opportunities in design, fabrication, and testing of integrated similar (ceramic-ceramic) and dissimilar (ceramic-metal) material systems will be discussed. Experimental results for bonding and integration of SiC based LDI fuel injector, high conductivity C/C composite based heat rejection system, solid oxide fuel cells system, ultra high temperature ceramics for leading edges, and ceramic composites for thermostructural applications will be presented. Potential opportunities and need for the development of innovative design philosophies, approaches, and integrated system testing under simulated application conditions will also be discussed.
Ceramic Integration Technologies for Aerospace and Energy Systems

Technical Challenges and Opportunities

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Overview

• Introduction and Background
  – Global Energy Issues and Role of Ceramics
• Technical Challenges in Integration
  – Ceramic-Metal Systems
  – Ceramic-Ceramic Systems
• Ceramic Integration Technologies
  – High Temperature Systems: Thermostructural components
  – Energy Efficiency: MEMS-LDI Fuel Injector, Thermal Management (Heat Exchangers and Recuperators, etc.)
  – Alternative Energy: SOFC Systems
• Concluding Remarks
• Acknowledgments
Critical Role of Advanced Ceramic Technologies
in Energy and Aerospace Applications

- **Energy Production**
  - Fuel Cells, Thermoelectrics, Photovoltaics
  - Nuclear, Wind, Biomass

- **Energy Storage and Distribution**
  - Batteries, Capacitors, Hydrogen Storage Materials
  - High Temperature Superconductors

- **Energy Conservation and Efficiency**
  - Ceramic Components (Gas Turbines, Heat Exchangers, etc.), Coatings, Bearings

Vision of Future Aircraft and Engine Configurations
[Broichhausen (2005), Szodruch (2005)]

Hydrogen Intercooler/Recuperator Solar/Fuel Cells

Lower & Slower

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

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

Common Issues
Ceramic Integration Technologies

• Joining/Bonding
• Attachments, Fasteners/Rivets
• Repair

Affordable, Robust Ceramic Joining Technology
(ARCJoinT)

Apply Carbonaceous Mixture to Joint Areas
Cure at 110-120°C for 10 to 20 minutes

Apply Silicon or Silicon-Alloy (paste, tape, or slurry)
Heat at 1250-1425°C for 10 to 15 minutes

Advantages
• Joint interlayer properties are compatible with parent materials.
• Processing temperature around 1200-1450°C.
• No external pressure or high temperature tooling is required.
• Localized heating sources can be utilized.
• Adaptable to in-field installation, service, and repair.

1999 R&D 100 Award
2000 NorTech Innovation Award
ARCJoinT can be Used to Join a Wide Variety of Ceramic and Composite Materials

SiC-Based Ceramics
- Reaction Bonded SiC
- Sintered SiC
- CVD SiC, Porous SiC

SiC/SiC Composites
- Melt Infiltrated SiC/SiC
- CVI SiC/SiC Composites
- PIP SiC/SiC Composites

C/SiC Composites
- Melt Infiltrated C/SiC
- CVI C/SiC Composites
- PIP C/SiC Composites

C/C Composites
- CVI C/C Composites
- Resin Derived C/C
- C-C with MI

- Composites with Different Fiber Architectures and Shapes
- Ceramics with Different Shapes and Sizes

Effect of Surface Roughness on the Shear Strength of Joined CVI C/SiC Composites

CVI C/SiC Composites

Joints with As-Fabricated Surfaces
Joints with As-Fabricated/Machined Surfaces
Joints with Machined Surfaces
Microstructure of As-Fabricated and Joined CVI C/SiC Composites

CVI C/SiC Composites (as fabricated)

(both surfaces as received)

(both surfaces machined)

(one surface machined and one surfaces as received)

Specimen Geometry and Test Fixture Used for Compression Double-Notched Shear Tests

ASTM C 1292-95a (RT) and ASTM C 1425-99 (HT)

Specimen Dimensions

- Specimen length (L) : 30 mm (±0.10 mm)
- Distance between notches (h) : 6 mm (±0.10 mm)
- Specimen width (W) : 15 mm (±0.10 mm)
- Notch width (d) : 0.50 mm (±0.05 mm)
- Specimen thickness (t) : adjustable
Typical Stress-Strain Behavior Obtained During the Compression Double-Notched Shear Tests

- Shear strength of joints increases with temperature and is higher than the CVI SiC composite substrate.
- No apparent influence of surface condition on the shear strength of joints.
Integration of Advanced CMC-CMC Systems

High Temperature CMC Joining

CMC Control Surfaces

Hyper-X Program Leading Edge

Joining of CMCs for Ground Based Applications

LEA CMC Cooled Combustion Chamber

Development at Snecma and Onera through PWR & AFRL, A3CP program-tested at AFRL, Onera, and other test facilities

Joined C-C/SiC Intake Flap for HYTEX-German Hypersonic Technology Program (DLR)
Ceramic Integration Technologies:
Attachment Systems/Fasteners

C/SiC Nosecap and Skirts
(DLR/Astrium and MT)

C/SiC body Flaps (MT Aerospace)

NASA-DLR-MT Aerospace (X-38 Program)

New CMC Attachment Design Concepts
being developed at THALES
ALENIA SPACE ITALIA S.p.A. - TORINO

EXPERT CMC Open Flaps and
Attachments developed at MT Aerospace

CMC Shingle TPS Attachment (Snecma)
Glenn Refractory Adhesive for Bonding and Exterior Repair (GRABER)

Multiuse Capability/Versatility of GRABER
- Repair of cracks, gouges, small holes, and missing surface coatings
- Edge sealant/adhesive for Plug concept
- Gap filler for T-seals and other areas
- Sealing the edges, gaps, attachment areas for flexible ceramic/metallic wrap concepts for large area damage repair
- Prepregs made with various ceramic fabrics are useful for various high temperature applications in aerospace and ground based systems.

Post Test- Front Side  Post Test- Back Side

- 2005 R&D 100 Award
- Northern Ohio Live Magazine- Awards of Achievement, S&T Category- Runner Up

Bonding and Integration of MEMS-LDI Fuel Injector

Objective: Develop Technology for a SiC Smart Integrated Multi-Point Lean Direct Injector (SiC SIMPL-DI)
- Operability at all engine operating conditions
- Reduce NOx emissions by 90% over 1996 ICAO standard
- Allow for integration of high frequency fuel actuators and sensors

Possible Injector Approaches

1. Lean Pre-Mixed Pre-Evaporated (LPP)
   **Advantages** - Produces the most uniform temperature distribution and lowest possible NOx emissions
   **Disadvantages** - Cannot be used in high pressure ratio aircraft due to auto-ignition and flashback

2. Lean Direct Injector (LDI)
   **Advantages** - Does not have the problems of LPP (auto-ignition and flashback)
   - Provides extremely rapid mixing of the fuel and air before combustion occurs
Multi-Point Lean Direct Injector


Lean Direct Injector Fabricated by Bonding of SiC Laminates

SiC laminates can be used to create intricate and interlaced passages to speed up fuel-air mixing to allow lean-burning, ultra-low emissions

Key Enabling Technologies:
- Bonding of SiC to SiC
- Brazing of SiC to Metallic (Kovar) Fuel Tubes

Benefits of Laminated Plates
- Passages of any shape can be created to allow for multiple fuel circuits
- Provides thermal protection of the fuel to prevent choking
- Low cost fabrication of modules with complicated internal geometries through chemical etching
Leak Test of SiC Laminates Joined with Silicate Glass

Combustion air channels
Fuel holes
Leaks at the edge between joined laminates

Air should only flow through the fuel holes
Undesired leaks in the combustion air channels
Plugged fuel hole

Microprobe Analysis of Alloyed Ti Foil

Ti-6Al-4V (weight %)
Grey phase – Alpha alloy
White phase – Beta alloy

Microprobe from the cross-section of alloyed Ti foil (averages taken from several points near the edge and at the center of the foil)

<table>
<thead>
<tr>
<th>Phase</th>
<th>Atomic Ratio</th>
<th>Al</th>
<th>Fe</th>
<th>Ti</th>
<th>V</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Grey Phase</td>
<td>9.999</td>
<td>2.748</td>
<td>2.172</td>
<td>77.084</td>
<td>17.997</td>
<td>100.000</td>
</tr>
<tr>
<td>Weight (%)</td>
<td>5.999</td>
<td>0.051</td>
<td>0.042</td>
<td>86.774</td>
<td>2.988</td>
<td>100.000</td>
</tr>
<tr>
<td>Atomic Ratio White Phase</td>
<td>4.841</td>
<td>1.850</td>
<td>76.507</td>
<td>16.803</td>
<td>100.000</td>
<td></td>
</tr>
<tr>
<td>Weight (%)</td>
<td>2.748</td>
<td>2.172</td>
<td>77.084</td>
<td>17.997</td>
<td>100.000</td>
<td></td>
</tr>
</tbody>
</table>
Microprobe of α-SiC Bonded Using Ti Foil

Conditions: 1250 °C, 24 MPa, 2 hr, vacuum, 5 °C/min

Microcracking may be due to the formation of two detrimental phases:

- Phase B Ti₅Si₃CX – Ti₅Si₃ is highly anisotropic in its thermal expansion where CTE(c)/CTE(a) = 2.72 (Schneibel et al).
- Phase E – Ti₃Al has low ductility at low temperatures. Al can be in the range of 23-35 atm % (Djanarthany et al).

Both phases can contribute to thermal stresses and microcracking during cool down.


Secondary Electron Image of the Diffusion Bond - Alloyed Ti Foil and Trex CVD SiC
Diffusion Bonding of CVD-SiC Using PVD Ti Interlayer

20 Micron Ti Interlayer
Microcracking is still present due to the presence of $\text{Ti}_5\text{Si}_3\text{C}_x$.
Naka et al suggest that this is an intermediate phase.

10 Micron Ti Interlayer
No microcracking or phase of $\text{Ti}_5\text{Si}_3\text{C}_x$ is present.
Thin interlayers of pure Ti downslected as the preferred interlayer.

Phases in bond with the 20 µ Ti Interlayer – Atomic Ratios

<table>
<thead>
<tr>
<th>Phase</th>
<th>Ti</th>
<th>Si</th>
<th>C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Phase A</td>
<td>56.426</td>
<td>17.752</td>
<td>25.757</td>
</tr>
<tr>
<td>Phase B</td>
<td>35.794</td>
<td>62.621</td>
<td>1.570</td>
</tr>
<tr>
<td>Phase C</td>
<td>58.767</td>
<td>33.891</td>
<td>7.140</td>
</tr>
</tbody>
</table>

Phases in bond with the 10 µ Ti Interlayer – Atomic Ratios

<table>
<thead>
<tr>
<th>Phase</th>
<th>Ti</th>
<th>Si</th>
<th>C</th>
</tr>
</thead>
<tbody>
<tr>
<td>Phase A</td>
<td>56.621</td>
<td>18.690</td>
<td>24.686</td>
</tr>
<tr>
<td>Phase B</td>
<td>35.752</td>
<td>61.217</td>
<td>3.028</td>
</tr>
</tbody>
</table>

High Strength of Bonds Greatly Exceeds the Application Requirements

1” x 1” Bonded Substrates

Pull test tensile strengths:

- > 23.6 MPa (3.4 ksi)*
- > 28.4 MPa (4.1 ksi)*

* failure in the adhesive to the test fixture

1” Diameter Discs with a 0.65” Diameter Bond Area

Pull test tensile strengths:

- 13.4 MPa (1.9 ksi)
- 15.0 MPa (2.2 ksi)

Slightly higher strength from the highly polished SiC suggests that a smoother surface contributes to stronger bonds or less flawed SiC.

Failures are primarily in the SiC substrate rather than in the bond area.

The injector application requires a strength of about 3.45-6.89 MPa (0.5 - 1.0 ksi).
Three Part 10 cm (4") Diameter SiC Injector

Stacking Sequence
Top to Bottom

Top Surfaces

Small Fuel Holes

Bottom Surfaces

Large Air Holes

Fuel Swirler Detail

PVD Ti Coated

Detail Next Slide

Detailed View of the Thickest Injector Substrate
(~0.635 cm thick)

Air Hole Passage

Fuel Hole and Channel

www.nasa.gov
Integration Needs in Heat Rejection System

HRS Technologies

- Radiator Face Sheets
  - C/C Composites
  - CFRP Composites
- Saddle Materials
  - Foams
  - Composites (2D,3D)

Titanium

Bonding/Assembly
- Active Metal Brazing
- Adhesives

Testing and Analysis
- Lifetime Testing
- Property Database
- Performance database

Mechanical Attachments

Thermal Control Coatings and Treatments

Heat Pipes and Related Technologies

Brazed Radiator Sub-elements

Adhesive Bonded Radiator Sub-element
Thermal Expansion Mismatch Issues are Critical in Brazing of Metal-Composite System

Innovative joint design concepts, new braze materials, and robust brazing technology development are needed to avoid deleterious effects of thermal expansion mismatch.

Active Metal Brazing

- Ti tubes and plates brazed to P120 CVI C/C composite
- Several braze/solder compositions compared (processing Temp):
  - TiCuSil (910 C) foil and paste
  - CuSil-ABA (820 C) foil and paste
  - CuSin-1ABA foil (810 C)
  - Incusil foil (725 C)
  - S-Bond solder (~ 400 C)
- Two tests have proved successful:
  - Butt Strap Tension (BST)
  - Tube-Plate Tensile Test

- Require good wetting, bonding and spreading properties
- Desire minimal residual stress induced cracking in C/C
Microstructure of Brazed Ti and C-C Composites using TiCuSil and CuSil ABA Paste

Factors to consider:
- Braze composition, Processing variables
- Bonded area, Location of failure
- Architecture effects
Tube Tensile Test Data for Brazed Joints

![Graph showing test data for different brazing materials.]

Best spreading and largest bonded area

Failure Load, N

0 10 20 30 40 50 60 70

TCuSi Foil  TiCuSil Paste CuSil-ABA Foil CuSil-ABA Paste CuSil-1ABA Paste Incusil Foil

Failure Behavior of Ti Tube - C/C Composite Joints

![Images showing fracture surfaces of brazed joints.]

Tube and C/C plate fracture surfaces for CuSil-ABA paste braze material showing the bonded area of the outer ply of C/C brazed to the Ti tube (left) and C/C plate (right).

Tube and C/C plate fracture surfaces for Incusil ABA foil braze material showing the distinct bonded areas of the outer ply of C/C brazed to the Ti tube (left) and C/C plate (right).

Thermally-Induced Cracking in C/C Controls Shear Strength of Brazed Joints

For braze materials where there was strong bonding between the braze and the C/C and failure occurred in the outer- ply of the C/C

\[ \Delta \alpha = \alpha (\text{Ti}) - \alpha (\text{C/C}) \]

\[ \Delta T = T (\text{liquidus} \sim \text{processing}) - 25^\circ C \]

<table>
<thead>
<tr>
<th>Joint Material</th>
<th>Proc. Temp., C</th>
</tr>
</thead>
<tbody>
<tr>
<td>S-Bond</td>
<td>~ 400</td>
</tr>
<tr>
<td>CuSil ABA</td>
<td>830</td>
</tr>
<tr>
<td>TiCuSil</td>
<td>910</td>
</tr>
</tbody>
</table>

Brazed Structures with Woven K1100 and P120 C/C Facesheet/Foam/Titanium Tube

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

- Tube on flat: Lowest contact area, highest stress on joint
- Tube in shallow trough: Normal contact area, normal stress on joint
- Tube in deep trough: Highest contact area, lowest stress on joint

Tension test geometry

Shear test geometry
Locations of Potential Joint Failure in C/C Face Sheet/Poco Foam Saddle/Titanium

In addition the geometry of joining surfaces will affect strength of joint and influence spreading of joint material: flat to flat, flat to tube, curved surfaces…

Therefore, knowing the location of joint failure is critical

- Weakest link requiring further improvement
- Affects interpretation of results (material or test-dependent property)

Key factor: Bonded area dictated by braze composition and applied pressure, C/C constituent composition, fiber orientation, geometry of joined surface

Brazed Structures with Woven K1100 and P120 C/C Facesheet/Foam/Titanium Tube

Observations:

- The Poco HTC (carbon foam saddle material) is the weak link in the sandwich structures and not the brazed joint.
- Failure in tension and shear always occurs in the foam regardless of (1) P120 or K1100 woven face sheet materials or (2) whether the Ti tube was brazed to a curved Poco surface to maximize bond area or a flat Poco surface to maximize stress in the joint.
  - Maximum shear stresses subjected to braze exceeded 12 MPa based on load applied and approximate braze area.
  - Maximum tensile stresses subjected to braze exceeded 7 MPa based on load applied and approximate braze area.

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

- C-C composites were brazed to Cu-clad Mo using four ABA’s (Cu-ABA, Cusin, Ticusni and Ticusil).
- Good metallurgical bonding at joints, with some dissolution, diffusion, and solute redistribution.
- Ti preferentially segregated at the C-C/braze interface. Cu-ABA joints displayed the largest Ti concentrations at joint.
- Microhardness gradients exist at joint (peak HK is 300-350 in Cusin-1 ABA and Ticusil, and 200-250 HK in Cu-ABA and Ticusil).
- C-C/Cu-clad Mo systems are useful for thermal management applications.


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**Integration of YSZ/Steel for SOFC Applications**

- Gold-ABA and Gold ABA-V exhibit linear oxidation kinetics at 850 C. Gold-ABA-V shows faster oxidation kinetics than Gold-ABA.
- Ti in Gold-ABA and V in Gold-ABA V caused discoloration (darkening) of YSZ.
- The darkening is caused by Ti and V that act as oxygen getters and form oxygen-deficient YSZ. No reaction layers formed at joint.
- The oxygen-deficient YSZ is better wet by gold than stoichiometric YSZ.

Integration of YSZ/Steel for SOFC Applications

- Pd-base brazes (Palco and Palni) and Ag-base brazes (Palcusil-10 and Palcusil-15) were characterized for oxidation at 750°C.
- Structural changes accompany oxidation which is fastest for Palco, slowest for Palni, and intermediate for Palcusil-10 and Palcusil-15.
- All brazes were effective in joining yttria stabilized zirconia (YSZ) to stainless steel for solid oxide fuel cell (SOFC).
- Dissolution of YSZ and steel in braze, and braze constituents in YSZ and steel led to diffusion and metallurgically sound joints.
- Knoop hardness (HK) profiles are similar for all brazes, and exhibit a sharp discontinuity at the YSZ/braze interface.


Integration of YSZ/Steel for SOFC Applications

- Active braze alloys, Cu-ABA, Ticuni and Ticusil, were characterized for oxidation at 750-850°C.
- Oxidation is fastest for Ticusil, slowest for Cu-ABA, and intermediate for Ticuni.
- Brazes were used for joining yttria-stabilized-zirconia (YSZ) to stainless steel for Solid Oxide Fuel Cell (SOFC) applications.
- Interdiffusion, compositional changes, and reaction layer formation led to high-integrity joints.

Concluding Remarks

• Ceramic integration technologies are critically needed for the successful development and applications of ceramic components in a wide variety of energy and aerospace applications.
• Significant efforts are needed in developing joint design methodologies, understanding the size effects, and thermomechanical performance of integrated systems in service environments.
• Global efforts on standardization of integrated ceramic testing are required. In addition, development of life prediction models for integrated components is also needed.
• There have been a number of short term design, development, and evaluation efforts in various parts of the world. However, a concerted and long term sustained effort is needed to make the significant progress in this area.

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