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 (ΔCTE)</td>
<td>Residual stress (ΔCTE)</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>
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</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

Affordable and Robust Ceramic Joints with Tailorable Properties

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/SiC 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 machined)

(one surface machined and one surfaces as received)

Joined CVI C/SiC Composites (both 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

Compression Double Notch Shear Strength of Joined CVI SiC Composites at Different Temperatures

- 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

C-C Engine Valve  Attachments for Sensors

CMC Control Surfaces

Tube-panel sub-element

Hyper-X Program
Leading Edge

Integration of CMC-Metal and CMC-CMC System

LEA CMC Cooled Combustion Chamber

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

Jointed C-CSIC Intake
Flap for HYTEX-German Hypersonic Technology Program (DLR)

Joining of CMCs for Ground Based Applications

Heat Exchanger Substructure  Radial Pump Wheel

C-CSIC Composites
Ceramic Integration Technologies:
Attachment Systems/Fasteners

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

C/SiC body Flaps (MT Aerospace)

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
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.

Arc Jet Testing
- Front View
- Side View
- Post Test- Front Side
- Post Test- Back Side
- * Analogue RCC Plug Sealed with GRABER 5A Crack Sealant
- * Survived the ArcJet Testing at JSC
- * 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

(Left) Multi-Point Lean Direct Injector accelerates fuel-air mixing and has small recirculation zones with short residence time that reduces NOx emission.

(Center) 3-inch square metal MP-LDI with 45 injectors.

(Right) Detail of fuel and airflow.


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>Al</th>
<th>Fe</th>
<th>Ti</th>
<th>V</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Atomic Ratio</td>
<td>Grey Phase</td>
<td>10.196</td>
<td>0.042</td>
<td>86.774</td>
<td>2.988</td>
</tr>
<tr>
<td>Weight (%)</td>
<td>Grey Phase</td>
<td>5.999</td>
<td>0.051</td>
<td>90.632</td>
<td>3.318</td>
</tr>
<tr>
<td>Atomic Ratio</td>
<td>White Phase</td>
<td>4.841</td>
<td>1.850</td>
<td>76.507</td>
<td>16.803</td>
</tr>
<tr>
<td>Weight (%)</td>
<td>White Phase</td>
<td>2.748</td>
<td>2.172</td>
<td>77.084</td>
<td>17.997</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 \( \text{Ti}_5\text{Si}_3\text{C}_x - \text{Ti}_5\text{Si}_3 \) is highly anisotropic in its thermal expansion where \( \text{CTE}(c)/\text{CTE}(a) = 2.72 \) (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
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>SiC</td>
<td>0.011</td>
<td>54.096</td>
<td>45.890</td>
</tr>
<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>

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>17.792</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>

Microcracking is still present due to the presence of Ti₅Si₃Cₓ. Naka et al suggest that this is an intermediate phase.

No microcracking or phase of Ti₅Si₃Cₓ is present. Thin interlayers of pure Ti downs-selected as the preferred interlayer.

High Strength of Bonds Greatly Exceeds the Application Requirements

Pull test tensile strengths:
- > 23.6 MPa (3.4 ksi)*
- > 28.4 MPa (4.1 ksi)*

* failure in the adhesive to the test fixture

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

Detail of the Thickest Injector Substrate
(~0.635 cm thick)

Air Hole Passage
Fuel Hole and Channel
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

Mechanical Attachments

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

Heat Pipes and Related Technologies

Thermal Control Coatings and Treatments

Integration of Sub-elements for Heat Rejection System
Using Brazing and Adhesive Bonding

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

Mechanical Testing of Brazed/Soldered Joints

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

Failure Behavior of Ti Tube - C/C Composite Joints

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

\[ \Delta \alpha = \alpha_{(Ti)} - \alpha_{(C/C)} \]
\[ \Delta T = T_{(liquidus \sim processing)} - 25^\circ C \]

<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: Higher contact area, lowest 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

- Within C/C
- C/C – JM interface
- Within JM
- JM – Saddle interface
- Within Saddle
- Within Ti

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

- Excellent bonding of CuSil-ABA Braze to Poco HTC, C-C Composite, and Ti Tube
- Failure always occurred in Poco HTC (Saddle Materials)

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, Ticuni 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 Ticuni).
- C-C/Cu-clad Mo systems are useful for thermal management applications.


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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