Implementation Challenges for Sintered Silicon Carbide Fiber Bonded Ceramic Materials for High Temperature Applications

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Abstract
During the last decades, a number of fiber reinforced ceramic composites have been developed and tested for various aerospace and ground based applications. However, a number of challenges still remain slowing the wide scale implementation of these materials. In addition to continuous fiber reinforced composites, other innovative materials have been developed including the fibrous monoliths and sintered fiber bonded ceramics. The sintered silicon carbide fiber bonded ceramics have been fabricated by the hot pressing and sintering of silicon carbide fibers. However, in this system reliable property database as well as various issues related to thermomechanical performance, integration, and fabrication of large and complex shape components has yet to be addressed. In this presentation, thermomechanical properties of sintered silicon carbide fiber bonded ceramics (as fabricated and joined) will be presented. In addition, critical need for manufacturing and integration technologies in successful implementation of these materials will be discussed.
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Outline

• Introduction and Background
• Fiber Bonded Ceramics: Overview
  • Materials and Manufacturing
  • Microstructure (SEM, TEM)
  • Thermal Properties
• Key Implementation Challenges
  • Thermomechanical Performance
  • Integration Technologies
  • Robust Manufacturing and Cost
• Concluding Remarks
• Acknowledgments
Ceramic Matrix Composites (CMCs): 
*Past, Present and Future*?

- Tremendous potential for use of ceramic matrix composites in aerospace and ground-based applications.
- Many intrinsic advantages over other material classes.
- Unique capabilities relative to certain applications.
- Substantial, long-term government funding for research and development in these materials worldwide.
- But many scientific, technical, economic, and cultural problems *still remain* in wide scale use of these materials.

**Introduction/Background**

*Timescale for Development and Applications of CMCs*

- **1960** - Primitive Whisker/Metal Reinforcements
- **1970** - Key Theoretical Basis for CMCs
- **1980** - First CMCs with Fiber, Matrix & Interphase
- **1990** - Advances in Micromechanical Models and Failure Criteria
- **2000** - Woven/Braided Fiber Architectures
- **2015** - National/International Standards, Target Applications, Design Codes and Data Bases
- **Widespread Applications**
Introduction/Background

Although more complex and more expensive than monolithic ceramics, the added “toughness” of CMCs make them more inherently damage tolerant than monolithic ceramics.

(Fiber Bonded Ceramics Could Play a Key Role)

Monolithic Ceramics - brittle, catastrophic failure

Fiber Bonded Ceramics - tough, “graceful” failure

Composite (CMC) - tough, “graceful” failure

SiC Fiber Bonded Ceramics

Processing and Microstructure
Synthesis of Amorphous Si-Al-C-O Fiber

- Polyalumino-carbosilane
- Melt spinning @220°C
- Curing in Air @160°C
- Firing in Ar gas up to 1300°C
- Forming continuously to fiber shape
- Changing to ceramic fiber

Fabrication of SiC Fiber Bonded Ceramics (SA-Tyrannohex)

- Amorphous Si-Al-C-O fiber
- Cut & Lamination
- Hot Pressing at ~1900°C, 50MPa, 1h in Ar gas

T. Ishikawa, Ube Industries

www.nasa.gov
Forming Process of SA-Tyrannohex during Hot Pressing

1. Under high pressure & high temp. in a hot press
2. Deforming fibers & Evaporating SiO and CO gas
3. Closed-pack hexagonal columnar structure
4. Carbon diffusion from the center of fibers to its surface
5. Unique SA-Tyrannohex structure

T. Ishikawa, Ube Industries

Microstructure of Fiber Bonded Ceramics

- **Unique Microstructure**
  - Hexagonal columnar SiC polycrystalline fibers
  - 10-20 nm turbostratic carbon interfacial layers

- **Good Properties**
  - High density (~98-99%)
  - High thermal conductivity (33 W/mK @ 1600°C)
  - High strength sustained up to 1600°C
  - Flexural strength ~300 MPa
  - High fracture toughness (1200 J/m² @ RT)
SiC Fiber Bonded Ceramics

Thermomechanical Properties

Thermomechanical Evaluation of Flexural Stress Rupture Behavior *(time-to-failure)*

- Specimen dimensions: 3mm × 4mm × 50 mm
- Pre-heat treatment: @1400°C for 10h in air
- 1µm outer silica coating

Fractography of tested specimens in SEM
- Focused on the tensile side
Stress versus Lifetime Behavior for SA-Tyrannohex™ Tested up to 1150°C in air

No distinguishable difference existed in the stress-dependence

Stress versus Lifetime Behavior for SA-Tyrannohex™ Tested up to 1400°C in Air

The time-to-failure ≥ 1300°C exhibited a stress-dependence
Stress versus Lifetime Behavior of MI SiC/SiC and SA-Tyrannohex up to 1300°C in Air

Fractography of SA-Tyrannohex after the Rupture Tests between 700-1400°C
SiC Fiber Bonded Ceramics  
*Joining and Integration Technologies*

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**Critical Needs for Integration Technologies**

- A wide majority of CMC components have to be integrated with existing metallic constituents or components either during component manufacturing or in service.
- It is important to understand the technical issues among the different material systems.
- Robust integration technologies can also play a key role in manufacturing of large size components (beyond existing manufacturing capabilities) utilizing building block approach.
- Building Block approach has been used through the ages and currently quite effectively in metal, polymer, and electronic industry.
Active Metal Brazing of Fiber Bonded Ceramics

- **Materials**
  - SA-Tyrannohex with two orientations
    - Parallel type
    - Perpendicular type
  - AgCuTi brazing alloy
    - Cusil-ABA
    - Ticusil

- **Conditions**
  - Temperature: 10-15°C above liquidus
  - Hold time: 5 min
  - Vacuum level: 10^-6 Torr

- **Procedure**

<table>
<thead>
<tr>
<th>Name</th>
<th>Composition (wt%)</th>
<th>$T_L$ (K)</th>
<th>$T_S$ (K)</th>
<th>CTE ($\times 10^{-6} \text{K}^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cusil-ABA</td>
<td>63.0Ag-35.25Cu-1.75Ti</td>
<td>1088</td>
<td>1053</td>
<td>18.5</td>
</tr>
<tr>
<td>Ticusil</td>
<td>68.8Ag-26.7Cu-4.5Ti</td>
<td>1173</td>
<td>1053</td>
<td>18.5</td>
</tr>
</tbody>
</table>

**Characterization**
- Mounted in epoxy, and polished
- Optical microscopy
- SEM-EDS

Uniform joint microstructure and good bonding
Joining of SA-Tyrannohex Material to Itself using the AgCuTi Brazing Alloy (Cusil-ABA & Ticusil)

Despite orientation of fiber and Ti content, microstructures of the joints were very similar.

Microstructure of the Joint Interfaces

- **Interlayer of SA-THX/Ticusil**
- **EDS mapping result**

<table>
<thead>
<tr>
<th>Pos.</th>
<th>Al</th>
<th>Si</th>
<th>Ti</th>
<th>Cu</th>
<th>Ag</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>16.0</td>
<td>84.0</td>
</tr>
<tr>
<td>2</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>58.5</td>
<td>1.5</td>
</tr>
<tr>
<td>3</td>
<td>0</td>
<td>39.0</td>
<td>50.1</td>
<td>10.9</td>
<td>0</td>
</tr>
<tr>
<td>4</td>
<td>0</td>
<td>8.0</td>
<td>87.9</td>
<td>4.1</td>
<td>0</td>
</tr>
<tr>
<td>5</td>
<td>1.1</td>
<td>98.5</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

atomic %
Summary of Joint Microstructure of SA-THX Materials using AgCuTi Alloy Brazes

- **Schematic**

- **Effect of Ti on the thickness of TiC layer on SA-Tyrannohex**

<table>
<thead>
<tr>
<th>Ti content</th>
<th>Orientation</th>
<th>Ti-Si region (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.75 wt%</td>
<td>Parallel</td>
<td>0.82 ± 0.20</td>
</tr>
<tr>
<td></td>
<td>Perpendicular</td>
<td>1.60 ± 0.68</td>
</tr>
<tr>
<td>4.5 wt%</td>
<td>Parallel</td>
<td>2.16 ± 0.88</td>
</tr>
<tr>
<td></td>
<td>Perpendicular</td>
<td>1.42 ± 0.45</td>
</tr>
</tbody>
</table>

- In the interface between SA-Tyrannohex and the metals, thin TiC layer and granular Ti-Si compound (could be Ti₅Si₃) were formed regardless of the orientation and Ti content.
- The joint microstructure of the SA-Tyrannohex using Ag-Cu-Ti alloy brazes was very similar regardless of the orientation and Ti content.

Procedure for Mechanical Evaluation

- **Joint Materials**
  - SA-THX(Δ& II)/Cusil-ABA

- **Single lap offset shear test**
  - Modified ASTM D905
  - R.T., 250, 650 and 750°C
  - Melting point: 815°C

- **Testing Machine**
  - Capacity: 22.5 kN
  - Crosshead speed: 0.5mm/min
  - Atmosphere: air
Mechanical Behavior SA-THX Joints as a Function of Test Temperature

Failed within SA-THX ≤ 250°C

Failed in joints ≥ 650°C

Shear Strength of the Perpendicular-Type SA-THX Joints as a Function of Test Temperature

The shear strength decreased with increase in temperatures.
Summary of Fractography

• Failure occurred between the filler metal and TiO₂ interaction layer
  - TiO₂ layer may be caused by oxidation of TiC, which was formed by the reaction between Ti in the filler metal and C in SA-Tyrannohex during the brazing.

• There are micro-cracks in SA-Tyrannohex in the vicinity of the interaction phase.
  - The micro-cracks could be brought about by degradation of SA-Tyrannohex strength, which would be caused by C migration in SA-Tyrannohex to form TiC.

• Since Cu reacts with O₂, CuO and Cu₂O phases were formed during the cooling after the test at 750°C.

Ceramic Joining and Integration
- Joining with Si-Hf Eutectic Phase Tape

Joining with Eutectic Phase Tapes (1 layer and 2 layers)
• SA-Tyrannohex (parallel) and SA-Tyrannohex (perpendicular)
• Joined with 2 mm offset for mechanical tests.
• Testing at R.T., 700°C and 1200°C
SiC Fiber Bonded Ceramics

Long Term Challenges

Environmental Issues in Manufacturing and Product Life Cycle Management

- ISO 14001 and ISO 14004 deal with Environmental Management System (EMS) - 1996.
- EMS provides a framework for an organization to manage the impact of its activities on the environment.
- Provides tools to help companies realize their own environmental policies, objectives, and targets.
- In Europe, European Community (EC) has established an Eco Management and Audit Scheme (EMAS) in 1997.

In Global Economy, Consumer Demand of High Quality Products with Low or No Environmental Impact, Standards Will Play Major Role
Performance vs Cost Issue

• It is quite clear that CMC industry is in a real dilemma
  - CMC users (customers) demand performance at cost, but cost is typically driven by market volume.
  - Small market volume means high cost and small number (or no) customers.
• Users (customers) are willing to pay the COST if the CMC is truly enabling.

Concluding Remarks

• Fiber Bonded Ceramics have a lot of potential for niche high temperature applications but the manufacturing processes are still evolutionary. Their use has been limited due to limited manufacturing base and cost.

• For the wide scale applications of these materials, reliable processes and properties have to be demonstrated at various levels (coupons to full scale components). In addition, multiscale modeling tools have to be developed and effectively utilized.

• The CMC community has to leverage their resources and make a concerted effort in finding out multiple applications and educating customers. High market volume will drop the cost and will be able to sustain the supplier base.
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