TEM Analysis of Diffusion-Bonded Silicon Carbide Ceramics Joined Using Metallic Interlayers

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1. Introduction
   properties and applications of SiC (SA-THX)
   purpose of diffusion bonding

2. Sample preparations used for diffusion bonding
   Substrates : SA-Tyrannohex™ (SA-THX)
   Interlayers : Ti-Mo, Ti-Cu foil

3. Experimental results
   STEM images of the bonding area
   TEM images and SAED patterns of the reaction compound

4. Discussion about the microstructure of the formed phases by diffusion bonding

5. Summary
SiC fiber-bonded ceramics, SA-Tyrannohex®

SiC composite material
1. Excellent mechanical properties
2. Good oxidation resistance
3. High thermal stability

Especially,

**SA-Tyrannohex (SA-THX)**
... SiC fiber-bonded ceramics

- High strength sustained **up to 1600°C in air**
- High fracture toughness **(1200 J·m⁻²)**

⇒ Promising material for high-temperature and extreme environment applications

E.g. injector applications, combustion liner, nuclear fusion reactor and turbine engine applications


For wide range uses of SA-THX

However, geometrical limitations hinder the wide use of SA-THX. It is difficult to fabricate large, or complex shaped components by Hot Pressing or CVD.

Therefore, new advanced methods are needed.

Under those circumstances,

One cost-effective solution for fabricating large, complex-shaped components is the joining of simple shaped ceramics.

In this study, we are going to focus on diffusion bonding.
Diffusion Bonding of SA-THX using metallic Interlayers

Used sample:
- Substrate: SA-THX SiC fiber-bonded ceramics, UBE Industries
- Metallic interlayer: Ti-foil, Mo-foil and Cu-foil, Goodfellow Corporation

Bonding process:
- Hot-press in 1200°C, 4 hour, vacuum 30MPa

Let’s Try with TRI!

Diffusion Bonding of a SA-THX using metallic Interlayers

Metallic Interlayer: **Ti-Mo** foil

Metallic Interlayer: **Ti-Cu** foil

Knoop hardness of diffusion bonded joints.

<table>
<thead>
<tr>
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<th>Average HK (∥ joint)</th>
<th>Average HK (∥ joint)</th>
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<tbody>
<tr>
<td>SA-THX/Ti/Mo/Ti/SA-THX</td>
<td>717.7 ± 273.6</td>
<td>758.9 ± 299.3</td>
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<tr>
<td>SA-THX/Ti/Cu/Ti/SA-THX</td>
<td>816.5 ± 43.9</td>
<td>—</td>
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<tr>
<td>SA-THX (un-bonded)</td>
<td>1244 ± 176</td>
<td>624 ± 205</td>
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</table>

M.C. Halbig, et. al., Ceramics International41(2015)2140–2149
Objectives

We diffusion bonded SiC and SiC (SA-THX and SA-THX) using Ti/Cu foil metallic interlayer. We carried out TEM and STEM observations with the diffusion bonded sample prepared by FIB technique.

1. Evaluate microstructures of the diffusion bonding area by TEM and STEM.

2. Characterize the reaction compound in the diffusion bonding area by STEM-EDS and SAED analysis.
Let's Try with TRI!

Experiment (FIB and STEM)

Focused Ion Beam, FIB
(Hitachi FB-2200)

Cs-corrected STEM
(Hitachi HD-2700)

Prepared thin samples for TEM and STEM.

Checked the thin samples prepared by FIB. Three-Observation mode:

SEM, BF-STEM and HAADF
Fabricating procedure of the thin sample (SIM image obtained by FIB)

1. Bonding area

2. W-depo (protection coating)

3. Protection coating

4. Final sample
STEM observation of the FIB sample

**Ti/Cu interlayer parallel to SiC fiber**

**Ti/Cu interlayer Perpendicular to SiC fiber**
- Only one layer in the bonding area
- No diffusion toward the side of SiC
- The reaction layer is composed of some grains and matrix around the grains.
Element Mapping obtained by STEM-EDS analysis

* FIB mesh: Cu metal

Cu-Si matrix + precipitated TiC grains?
TEM image and SAD patterns of Ti-C compound

Let’s Try with TRI!
TEM image and SAD patterns of Cu-Si compound

SAD pattern of $\text{Cu}_3\text{Si}$ : $\eta''$-phase (RT phase of $\text{Cu}_3\text{Si}$)

$//[001]$ 4 times superstructure $//[111]$ 3 times superstructure

M. Heuer, et. al., JAP 101, 123510 (2007)
TEM image and SAD patterns of Cu-Si compound

η-phase Cu$_3$Si:
(high temperature phase)
S.G.: P-3m1
a=0.4091 nm, b=0.7358

N. Mattern, et. al., HASYLAB ANNUAL REPORT (2001)
Consideration of the Results

1. RT (Before)
   - Ti-foil
   - Cu-foil

2. The top temperature 1200°C
   - Liquid-state (Cu-Ti-Si-C)

3. Cooling
   - TiC crystallization

4. RT (After)
   - TiC
   - Cu-Si compound

The bonding layer is composed of **TiC precipitations in Cu-Si matrix**.

**Cu-Si matrix** plays a role of the binder of TiC precipitations?
Consideration of the Results (CTE)

Metallic Interlayer: **Ti-Mo** foil

Metallic Interlayer: **Ti-Cu** foil

Coefficient of thermal expansion (CTE; $\alpha$)

<table>
<thead>
<tr>
<th>material</th>
<th>a-SiC</th>
<th>Mo</th>
<th>Ti</th>
<th>Cu</th>
<th>TiC</th>
<th>Ti$_3$Si5</th>
<th>Ti$_3$SiC$_2$</th>
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<tr>
<td>CTE $\alpha$ ($10^{-6}$K$^{-1}$)</td>
<td>3.2</td>
<td>5.1</td>
<td>8.4</td>
<td>16.8</td>
<td>7.4</td>
<td>6.1</td>
<td>16.6</td>
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<td>8.9</td>
<td>10</td>
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M.C. Halbig, et. al., Ceramics International 41(2015)2140–2149
Summary

1. We picked up thin samples from the bonded area of diffusion bonded SA-THX by a FIB micro-sampling technique. The prepared thin samples were sufficiently thin and less-damaged, and allowed the detailed evaluation by TEM and STEM.

2. The microstructure of diffusion bonded area was observed by STEM and TEM. The composition and crystal structures of the reaction compound were investigated by STEM-EDS and SAED method. The reaction layer of the diffusion bonding was composed of TiC precipitations in Cu-Si compound matrix.

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