

JOINING OF SILICON CARBIDE: DIFFUSION BOND OPTIMIZATION AND CHARACTERIZATION

Michael C. Halbig
Army Research Laboratory, NASA Glenn Research Center
Cleveland, OH, USA

Mrityunjay Singh
Ohio Aerospace Institute, NASA Glenn Research Center
Cleveland, OH, USA

ABSTRACT

Joining and integration methods are critically needed as enabling technologies for the full utilization of advanced ceramic components in aerospace and aeronautics applications. One such application is a lean direct injector for a turbine engine to achieve low NO_x emissions. In the application, several SiC substrates with different hole patterns to form fuel and combustion air channels are bonded to form the injector. Diffusion bonding is a joining approach that offers uniform bonds with high temperature capability, chemical stability, and high strength. Diffusion bonding was investigated with the aid of titanium foils and coatings as the interlayer between SiC substrates to aid bonding. The influence of such variables as interlayer type, interlayer thickness, substrate finish, and processing time were investigated. Optical microscopy, scanning electron microscopy, and electron microprobe analysis were used to characterize the bonds and to identify the reaction formed phases.



JOINING OF SILICON CARBIDE: DIFFUSION BOND OPTIMIZATION AND CHARACTERIZATION

Michael C. Halbig¹ and Mrityunjay Singh²

1 - U.S. Army Research Laboratory, Vehicle Technology Directorate, Cleveland, Ohio

2 - Ohio Aerospace Institute, Cleveland, OH



VEHICLE TECHNOLOGY DIRECTORATE



MS&T'08, Materials Science & Technology Conference & Exposition
Pittsburgh, PA, October 5-9, 2008.



Outline

- **Motivation and Application** – Micro-Electro-Mechanical Systems Lean Direct Injector (MEMS LDI) for Advanced Aircraft Gas Turbines
- **Joint Processing** - Diffusion Bonding With a Titanium Interlayer
- **Characterization** – Optical Microscopy, SEM, Microprobe
- **Near Term Plans** – Subcomponent Fabrication Issues and Further Diffusion Bond Optimization
- **Summary and Conclusions**



Motivation

Improve Ceramic to Ceramic and Ceramic to Metal Joining Methods

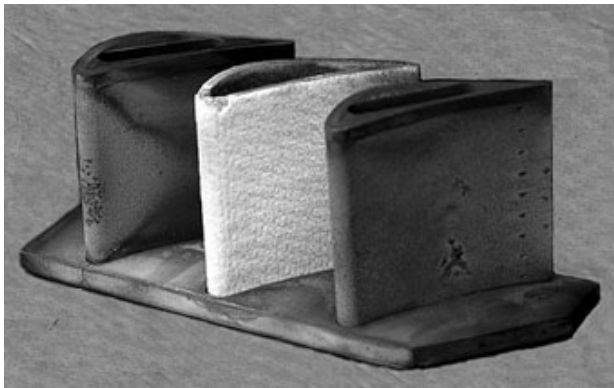
- allow for the fabrication of complex shapes and structures.
- allow for easier integration of ceramics components into engine systems.

(Ceramics can be either monolithics or fiber reinforced composites).

Examples of Component Applications Benefiting from Joining Technology

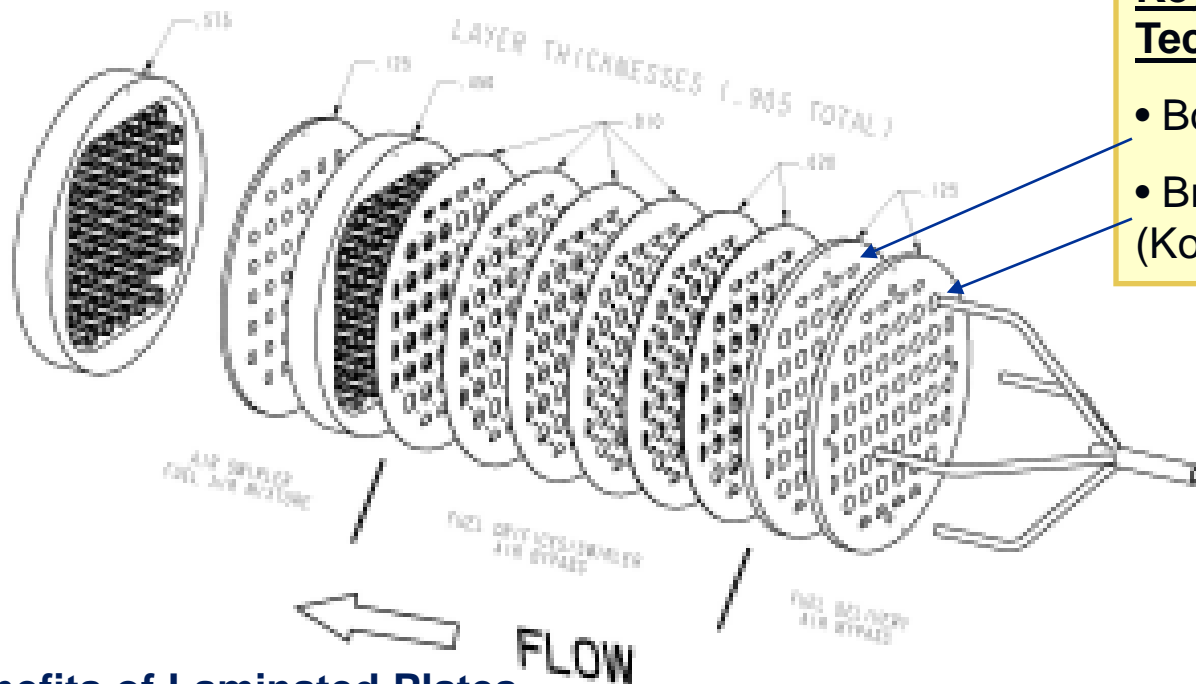
– components for aeronautic and ground based engine applications (i.e. ceramic turbine vanes, blades, injectors, rotors, combustor liners, valves, and heat shields) and fusion reactor components, optical space components, and bi-layer armor.

Benefits – simplified component fabrication, enabling technology, increased operating temperatures, improved efficiency, and reduced cooling requirements.



Fabrication of Lean Direct Injector Components by Diffusion 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



Current Approach of Joining SiC With a Ti Layer

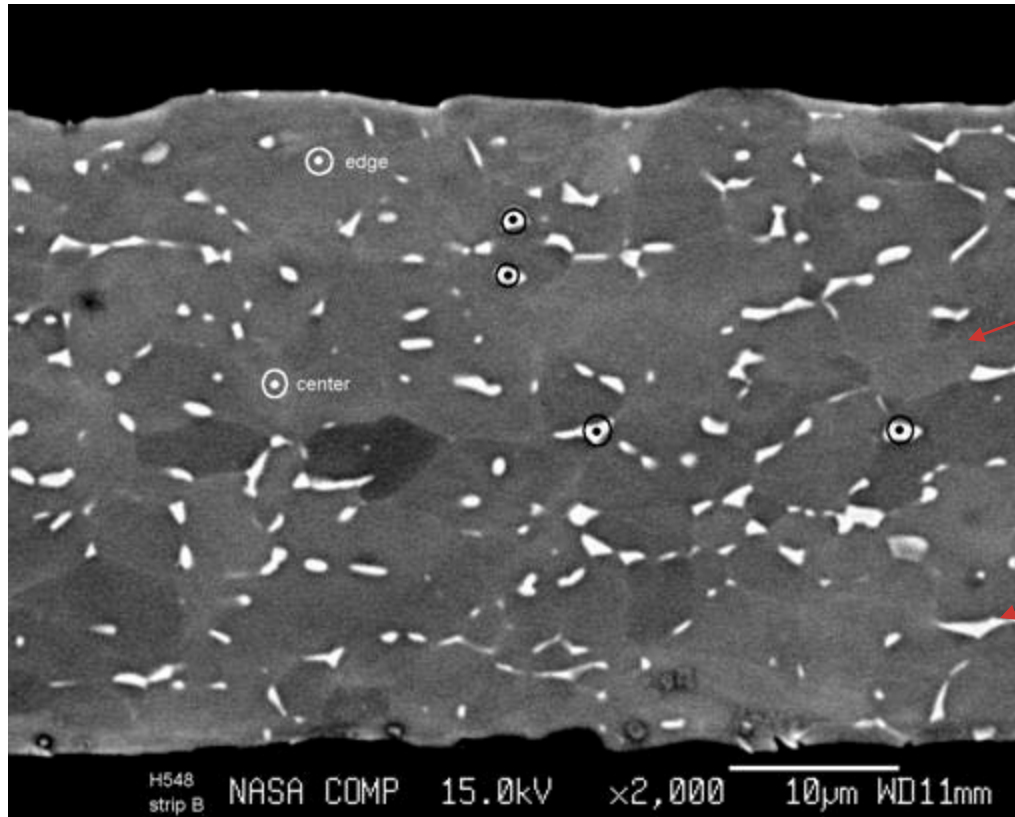
Advantages of Diffusion Bonding Using a Ti Layer

- Uniform Ti layers can be applied
- Ti can be applied by different methods (foil, PVD and other coating approaches)
- High strength and leak-free bonds
- Good high temperature stability

The objective is to develop joining technology that has the following capabilities necessary for the injector application:

- Joining of relatively large geometries (i.e. 4" diameter discs)
- Leak-free at an internal pressure of 200 psi (1.38 MPa)
- Stability and strength retention at 800°F (427°C)

Alloyed Ti Foil



Ti-6Al-4V (weight %)

Grey phase – Alpha alloy

White phase – Beta alloy

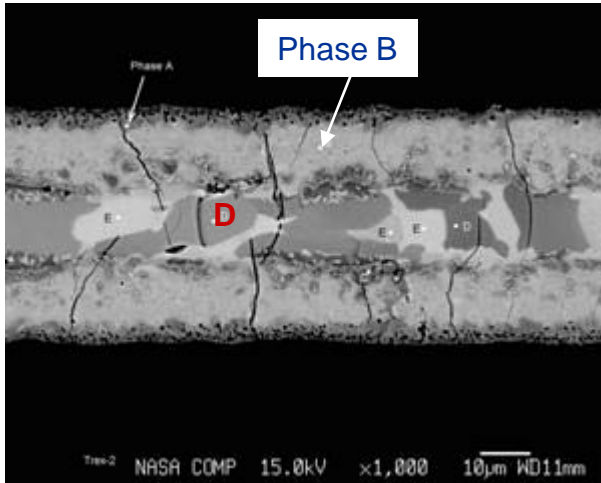
Microprobe from the cross-section of alloyed Ti foil (averages taken from several points near the edge and at the center of the foil)

| | Phase | Al | Fe | Ti | V | Total |
|--------------|-------------|--------|-------|--------|--------|---------|
| Atomic Ratio | Grey Phase | 10.196 | 0.042 | 86.774 | 2.988 | 100.000 |
| Weight (%) | Grey Phase | 5.999 | 0.051 | 90.632 | 3.318 | 100.000 |
| Atomic Ratio | White Phase | 4.841 | 1.850 | 76.507 | 16.803 | 100.000 |
| Weight (%) | White Phase | 2.748 | 2.172 | 77.084 | 17.997 | 100.000 |

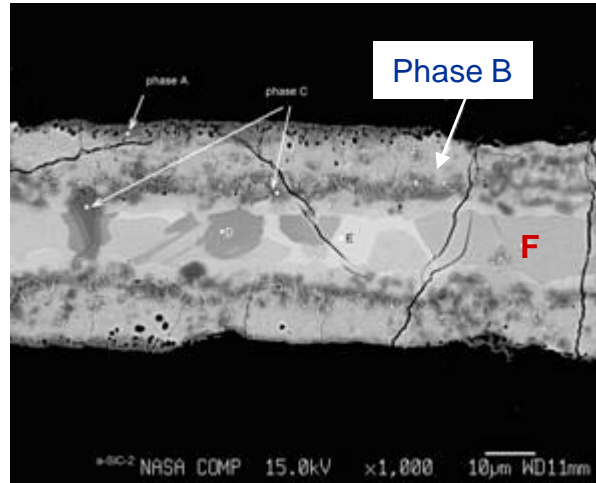
Bonding with the Alloyed Ti Foil Between Different SiC Substrates



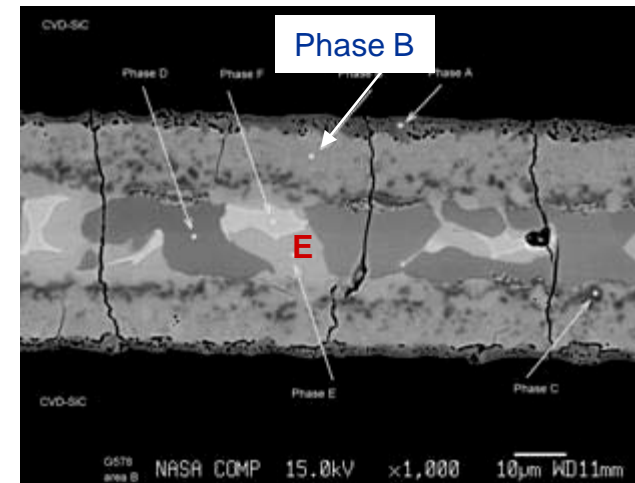
TREX CVD SiC



CRYSTAR α -SiC



Rohm & Haas CVD SiC



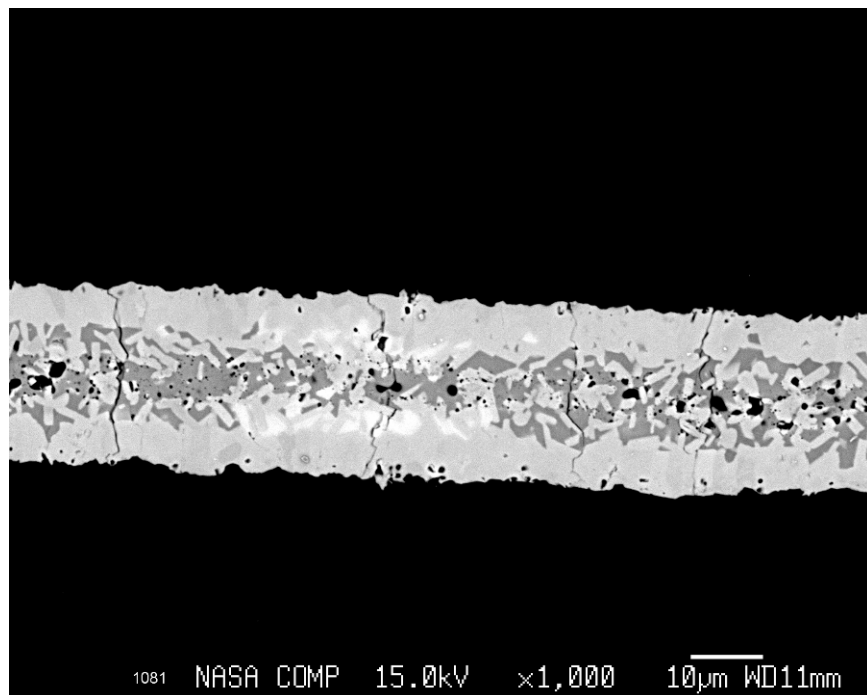
Microcracks formed regardless of the substrate and variations in the processing: higher temperatures, higher pressures, slower cooling rate.

Microcracking may be due to thermal stresses during cooling down from processing:

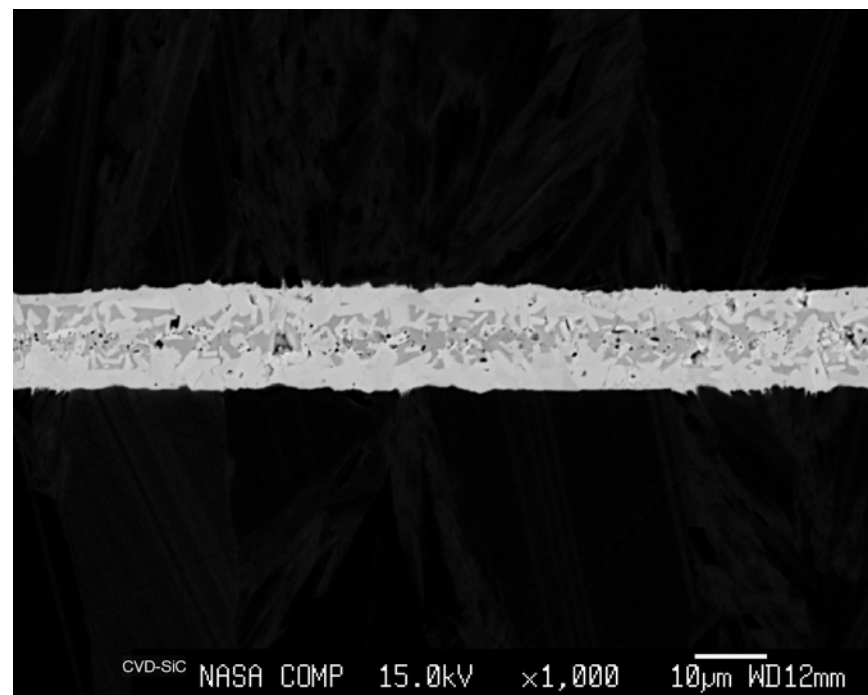
- Phase B (same in all three micrographs) - $Ti_5Si_3C_x$ (Ti_5Si_3) is highly anisotropic in its thermal expansion where $CTE(c)/CTE(a) = 2.72$ (Schneibel et al).
- Central core of diffusion bond has concentrated alpha and beta Ti alloy phases. The alpha phase has an anisotropic thermal expansion which is 20 % greater along the c-axis (Boyer, Welsch, and Colling). Also, the beta phase has a thermal expansion that is 6 x higher in the temperature range of 600-1000°C ($5.8 \times 10^{-5}/^\circ C$) compared the thermal expansion below 600°C ($9.2 \times 10^{-6}/^\circ C$) (Elmer et al).

Diffusion Bonds from Using PVD Ti as the Interlayer

20 Micron Ti Interlayer



10 Micron Ti Interlayer

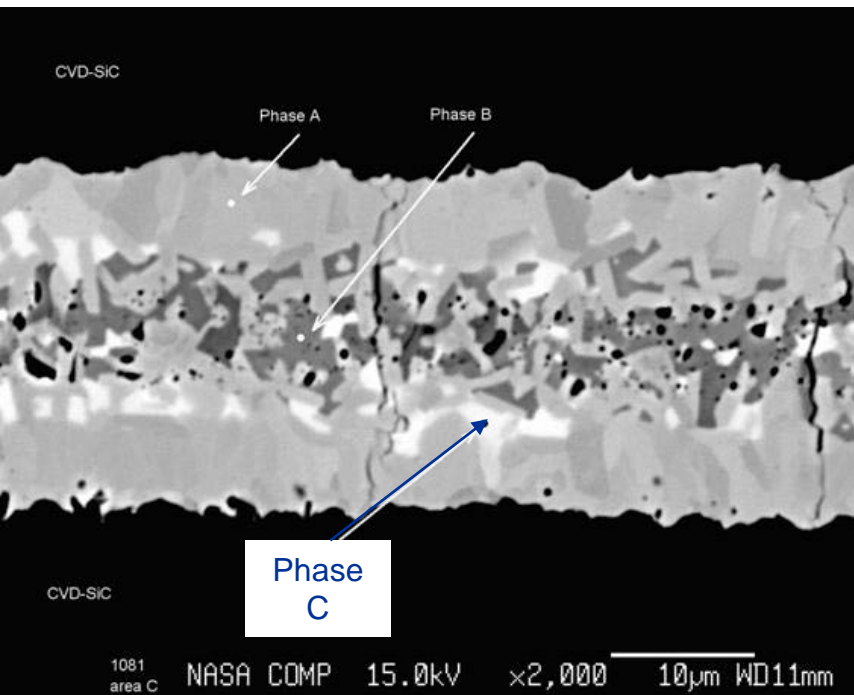


Diffusion Bonds from Using PVD Ti as the Interlayer

20 Micron Ti Interlayer

Microcracking is still present due to the presence of $Ti_5Si_3C_x$.

Naka et al suggest that this is an intermediate phase.



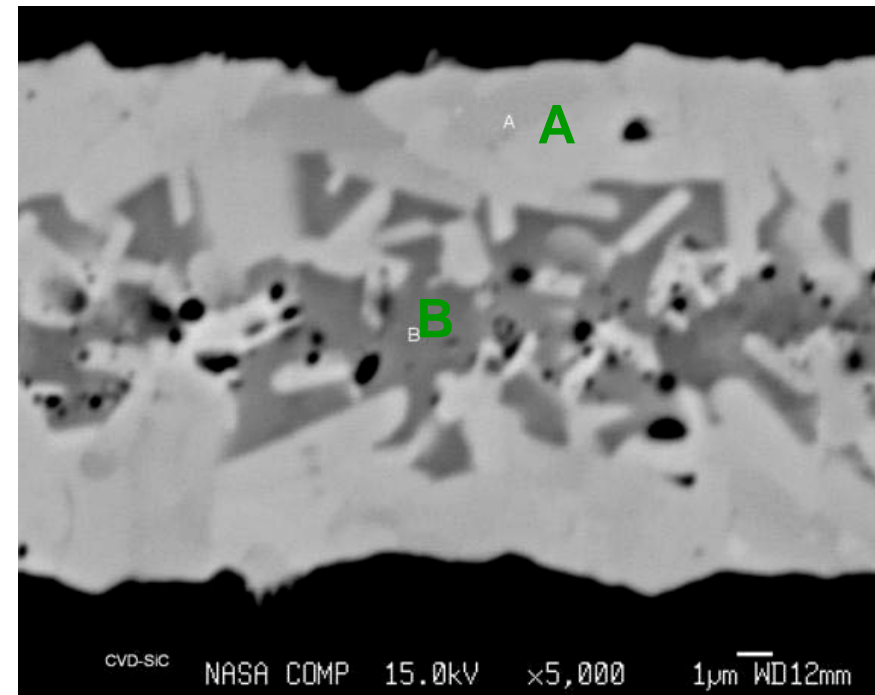
Phases in bond with the 20 μ Ti Interlayer – Atomic Ratios

| Phase | Ti | Si | C |
|---------|--------|--------|--------|
| Phase A | 56.426 | 17.792 | 25.757 |
| Phase B | 35.794 | 62.621 | 1.570 |
| Phase C | 58.767 | 33.891 | 7.140 |

10 Micron Ti Interlayer

No microcracking or phase of $Ti_5Si_3C_x$ is present.

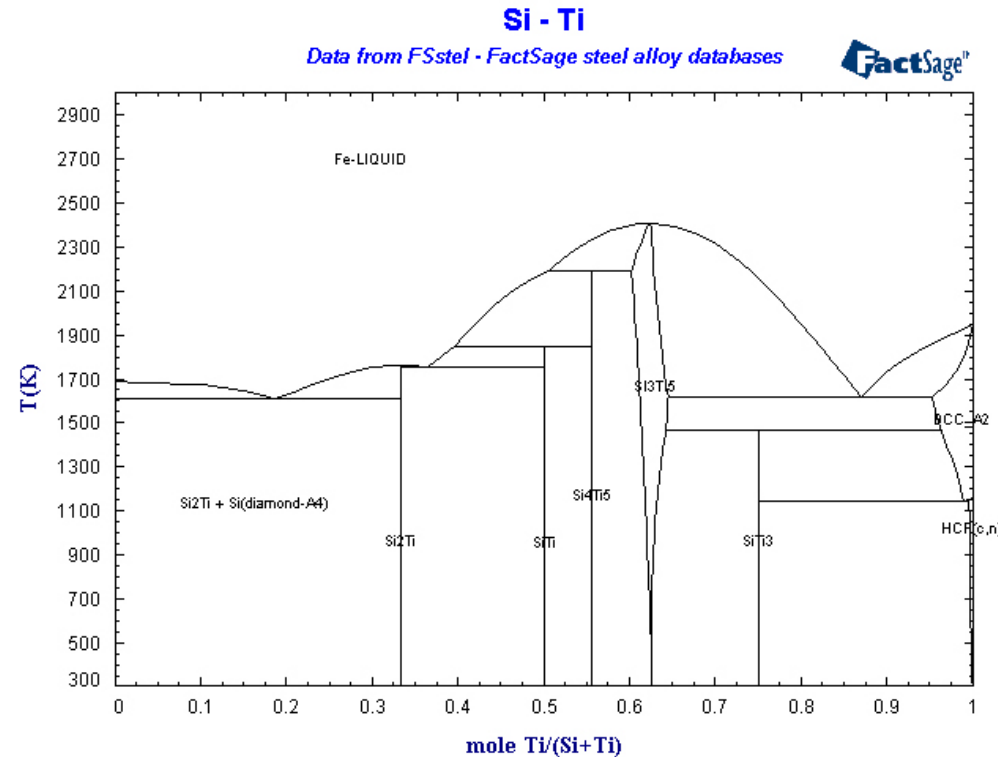
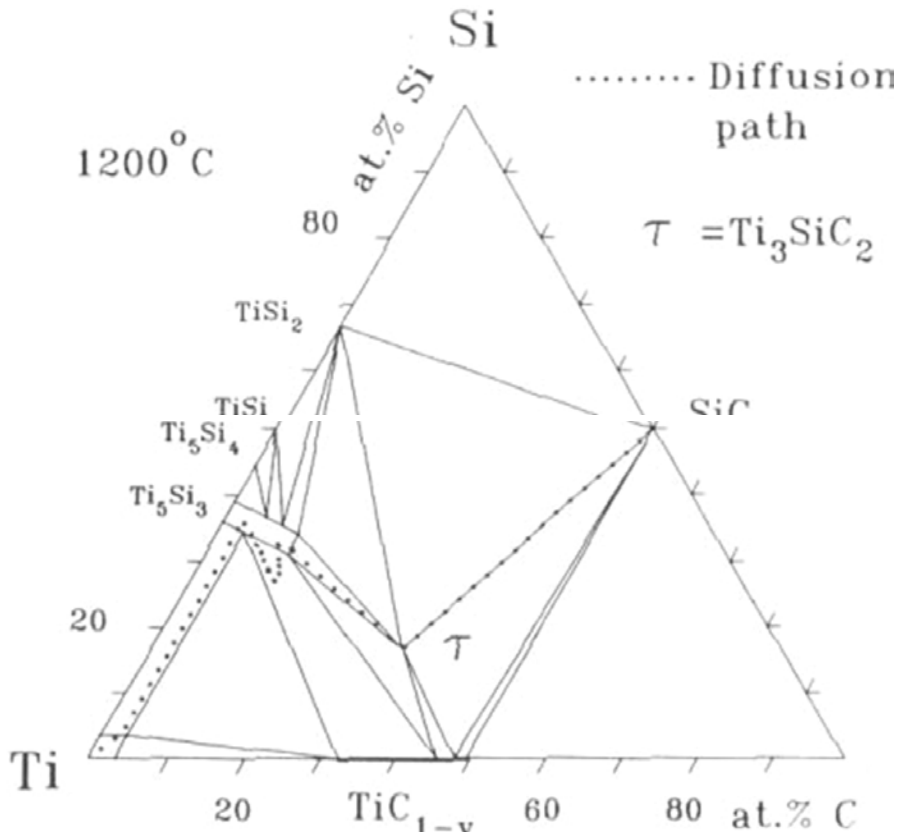
Thin interlayers of pure Ti down-selected as the preferred interlayer.



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

| Phase | Ti | Si | C |
|---------|--------|--------|--------|
| SiC | 0.011 | 54.096 | 45.890 |
| Phase A | 56.621 | 18.690 | 24.686 |
| Phase B | 35.752 | 61.217 | 3.028 |

The Phases of Ti-Si-C and the Intermediate Phase of Ti_5Si_3



An isothermal section of the Ti-Si-C phase diagram at 1200°C. The diffusion path is denoted by the dotted line. SiC/Ti₃SiC₂/Ti₅Si₃/two-phase Ti₅Si₃ plus TiC_{1-y}/Ti₅Si₃/Ti

F. Goesmann and R. Schmid-Fetzer, Semicond. **Sd. Technol.** **10** (1995) 1652-1658.



Effect of Ti Foil Thickness and Processing Time on the Resulting Diffusion Bonds - second processing matrix investigated

The following conditions were investigated:

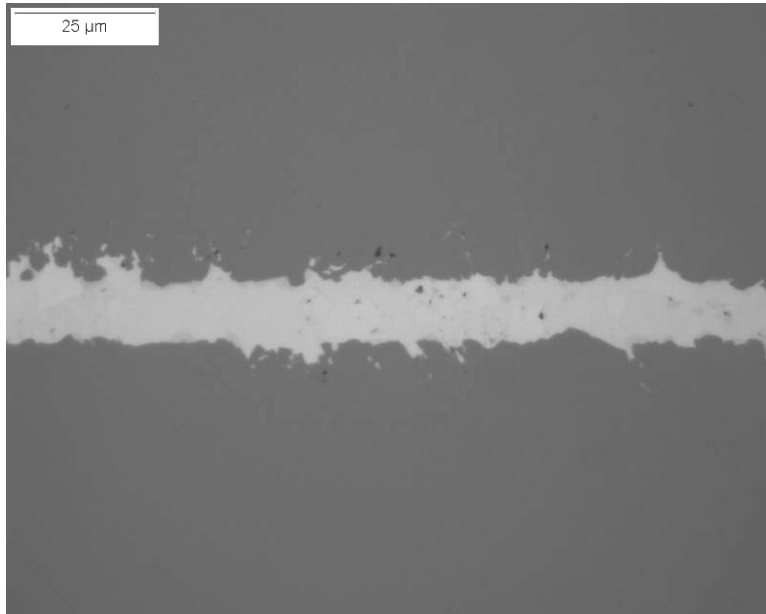
- 10 micron foil at 2 hr
- 20 micron foil at 1, 2, and 4 hr
- 50 micron foil at 1, 2, 4, 8 and 16 hr

* all other conditions are the same as before: 1250 °C, 30 MPa applied pressure, vacuum, 2 °C /min cool down

However, the processing temperature may have actually been as high as 1400 °C for all bonds.



SiC Substrates Joined with a 10 μm Ti Foil Interlayer for 2 hr

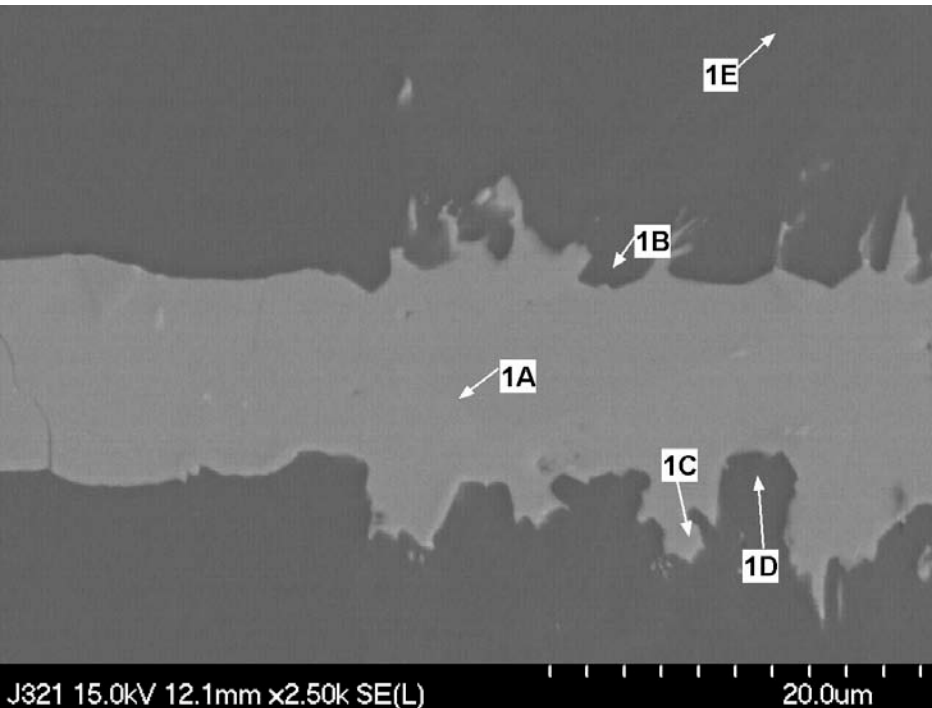


Similar results to the diffusion bond formed with a 10 micron thick PVD Ti coating

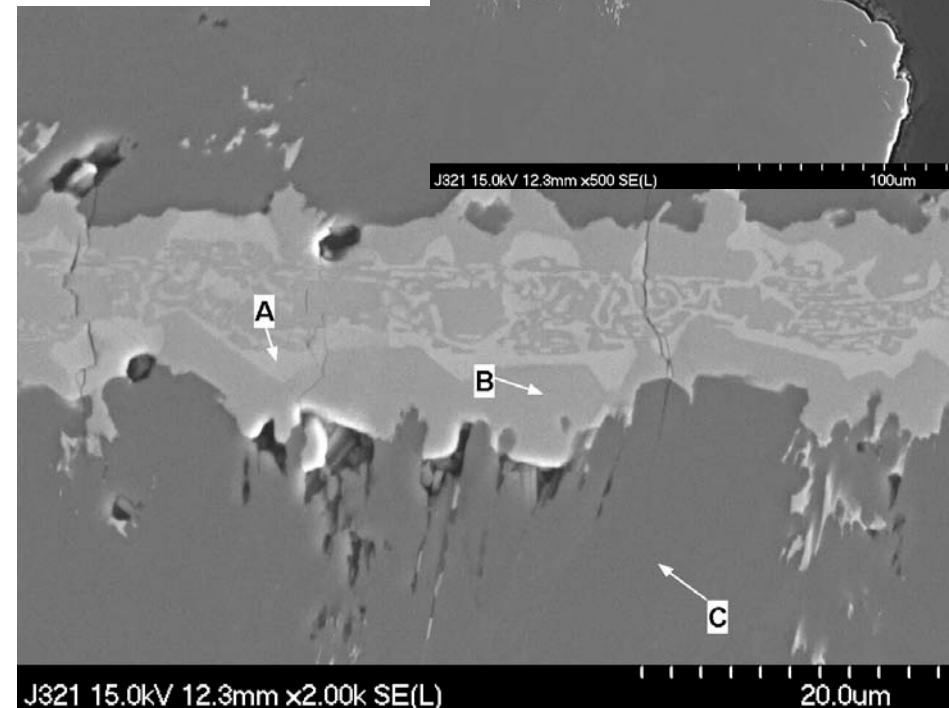
- no microcracks

SEM of SiC Substrates Joined with a 10 μm Ti Foil Interlayer for 2 hr

Center of the Bond Line



Near and at the Edge



Phases in atomic %

1A and 1C – C 49.7, Si 13.5, Ti 36.8

1B, 1D and 1E – C 56.8, Si 43.2

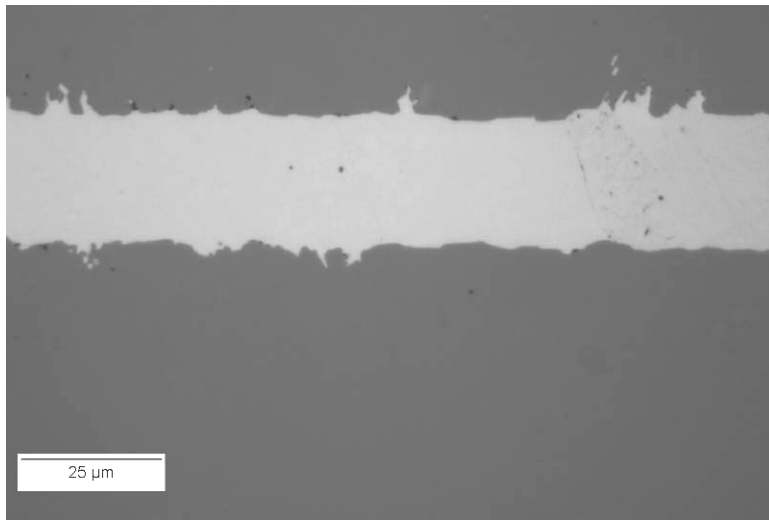
Phases in atomic %

A – C 50.2, Si 11.9, Ti 35.3, Fe 2.6

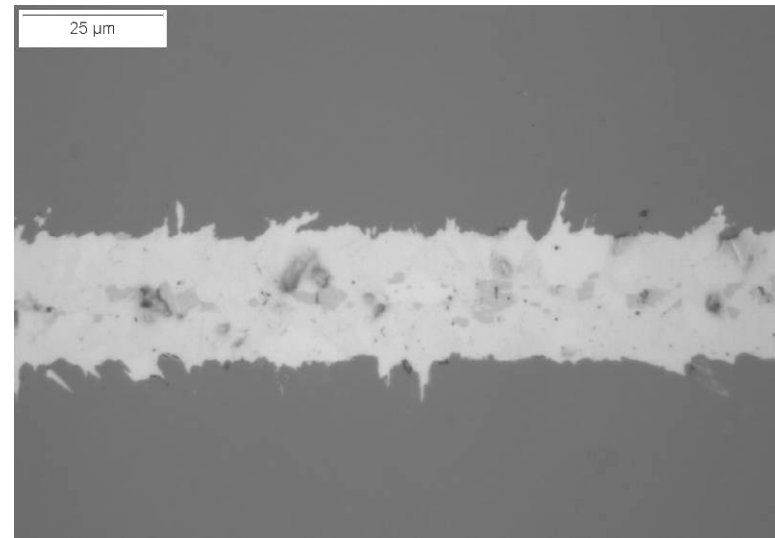
B – C 56.3, Ti 43.7

C – C 54.5, Si 45.5

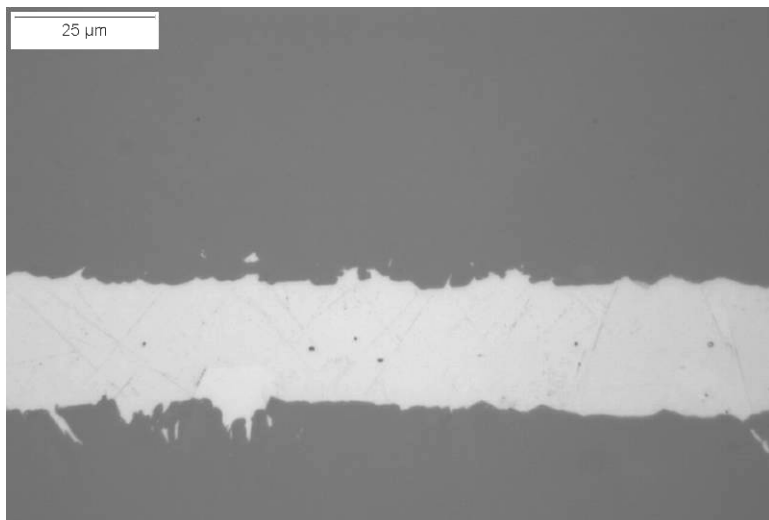
SiC Substrates Joined with a 20 μm Ti Foil Interlayer



1 hr.



2 hr.



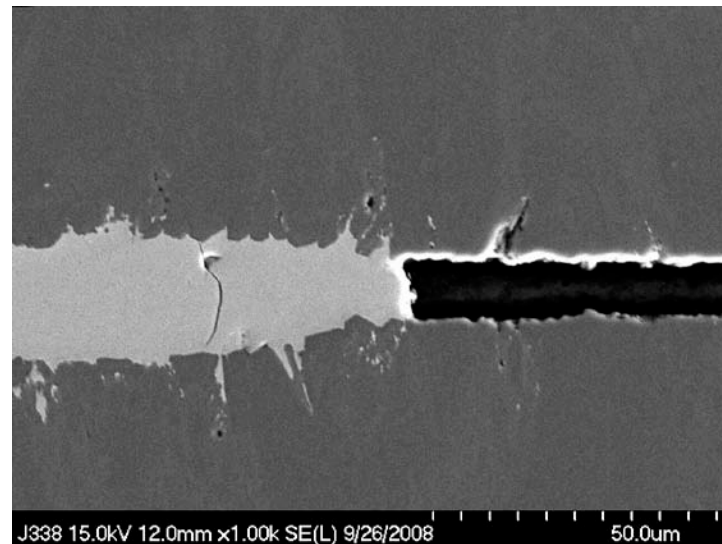
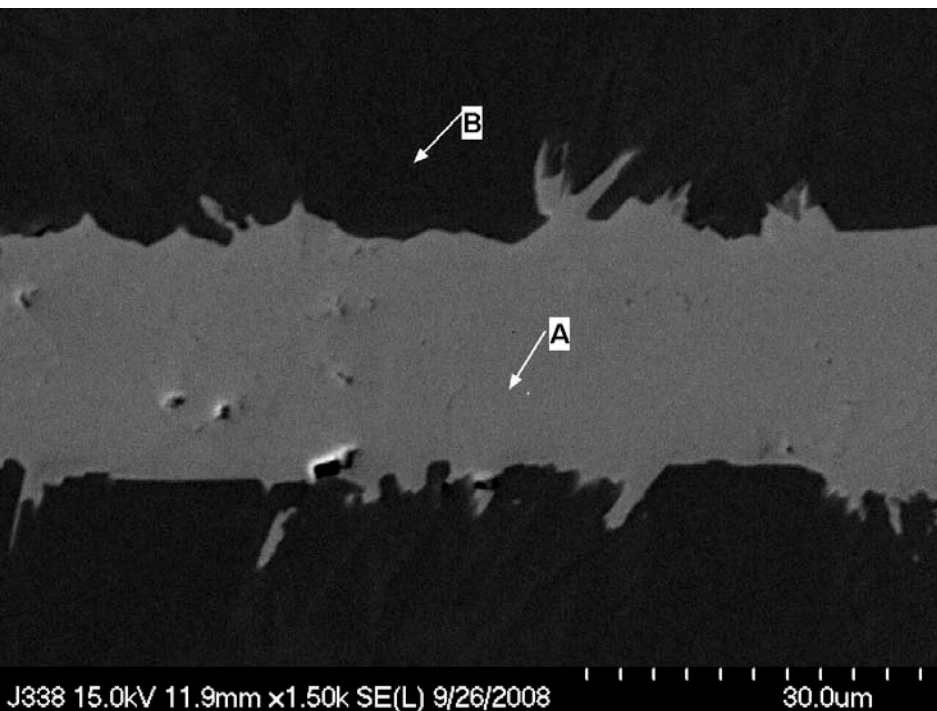
4 hr.

An intermediate phase is observed after 2 hr but not after 4 hr.

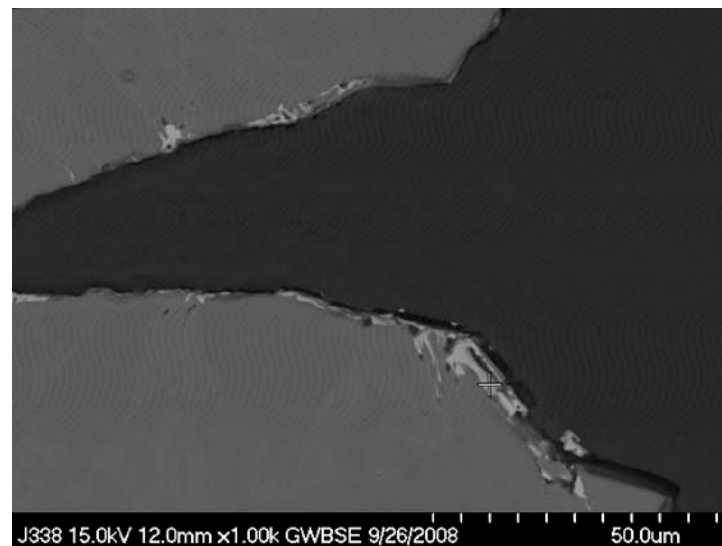
- minimal microcracking

SEM - SiC Substrates Joined with a 25 μm Ti Foil Interlayer For 4 hr

Center of the Bond Line



Near and at the Edge

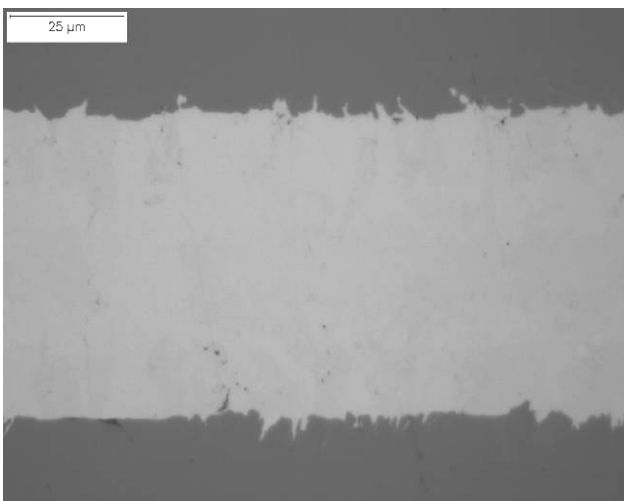


Phases in atomic %

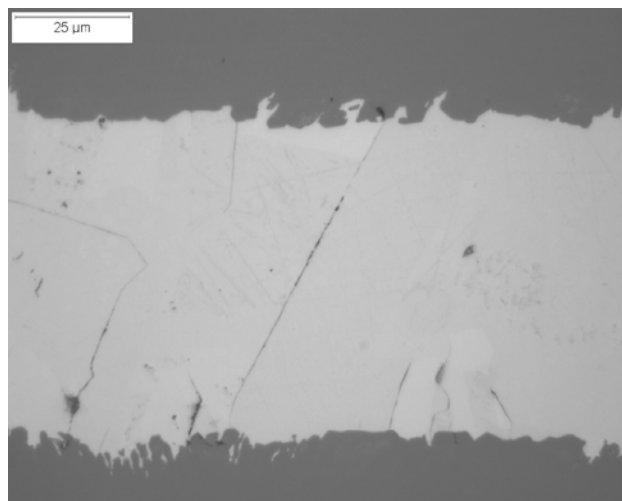
A – C 49.9, Si 13.4, Ti 36.7

B – C 57.6, Si 42.4

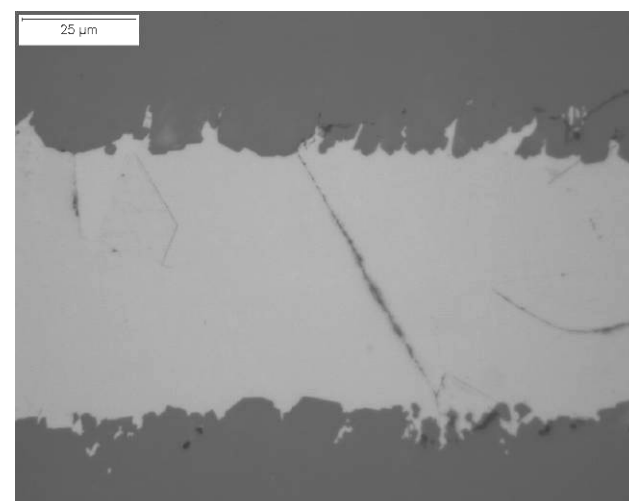
SiC Substrates Joined with a 50 μm Ti Foil Interlayer



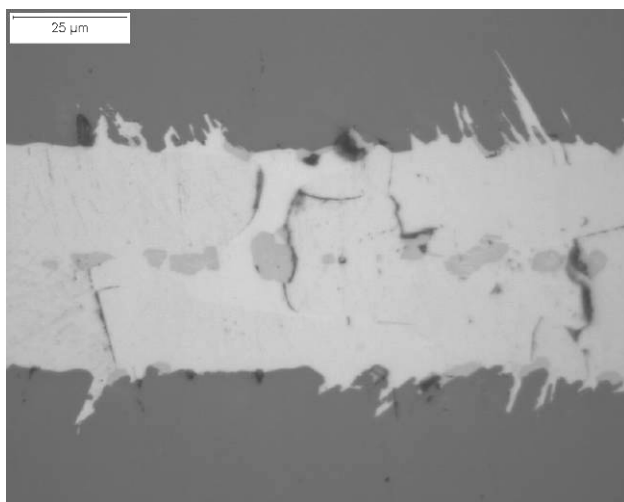
1 hr.



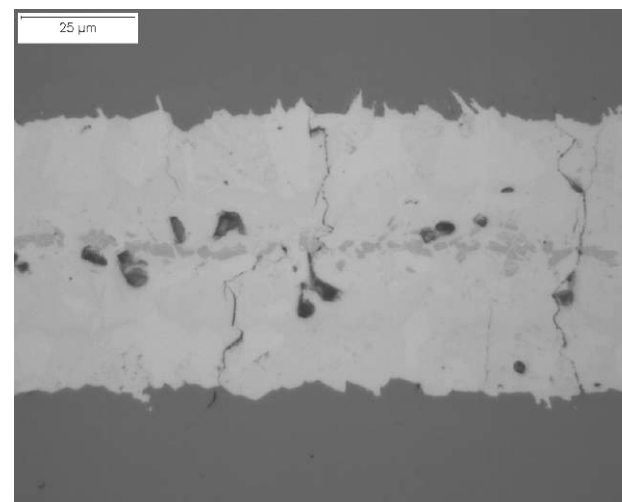
2 hr.



4 hr.



8 hr.

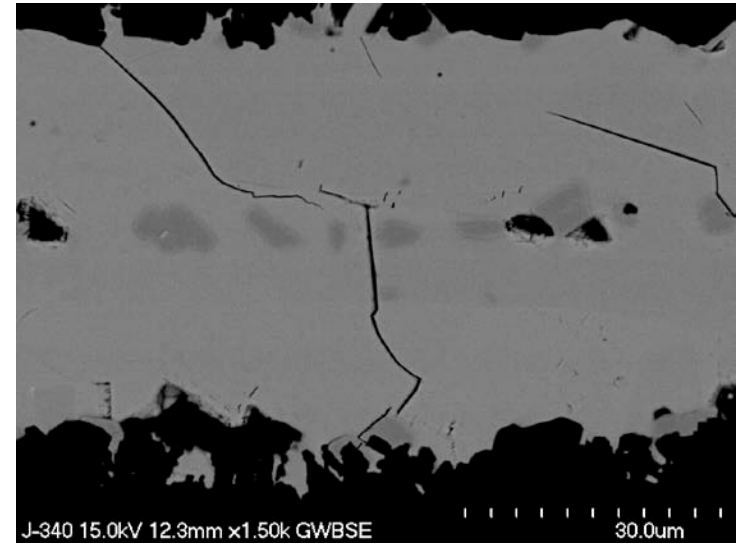
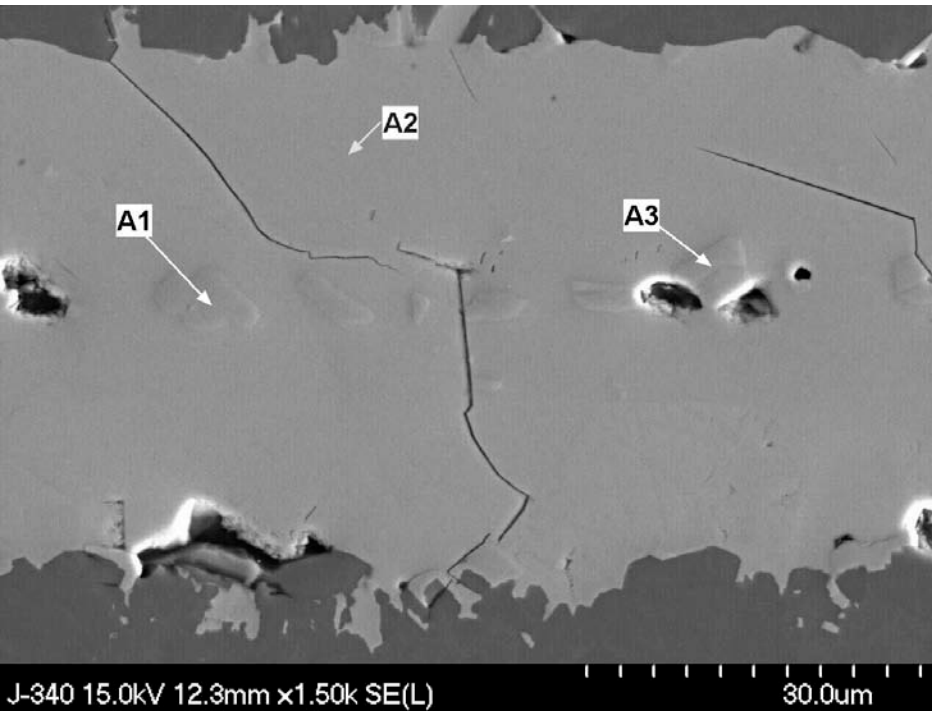


16 hr.

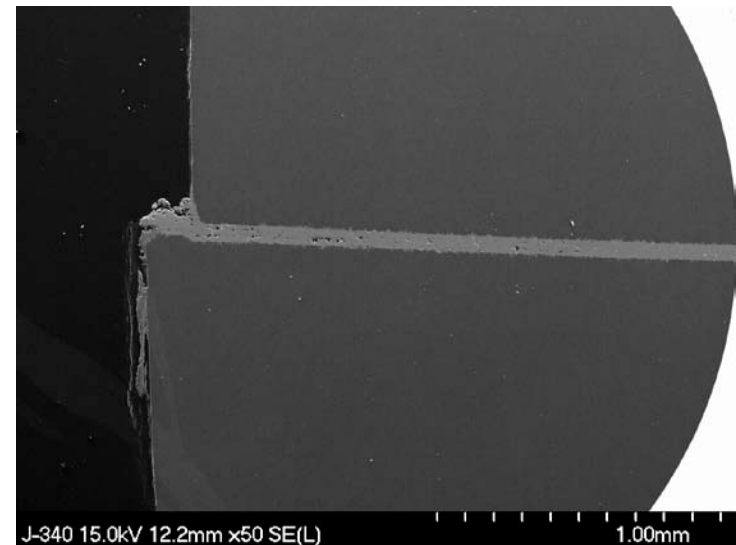
Over time, the formation of microcracks and an intermediate phase are observed followed by void formation and a lessening of the intermediate phase.

SEM - SiC Substrates Joined with a 50 μm Ti Foil Interlayer For 8 hr

Center of the Bond Line



Backscattered Image and Flow at an End



Phases in atomic %

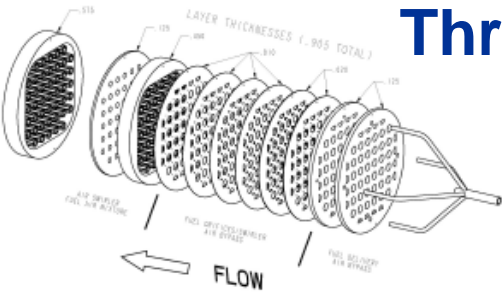
A1 – C 58.3, Si 1.2, Ti 40.5

A2 – C 54.0, Si 12.4, Ti 33.6

A3 – C 57.0, Si 3.3, Ti 39.7



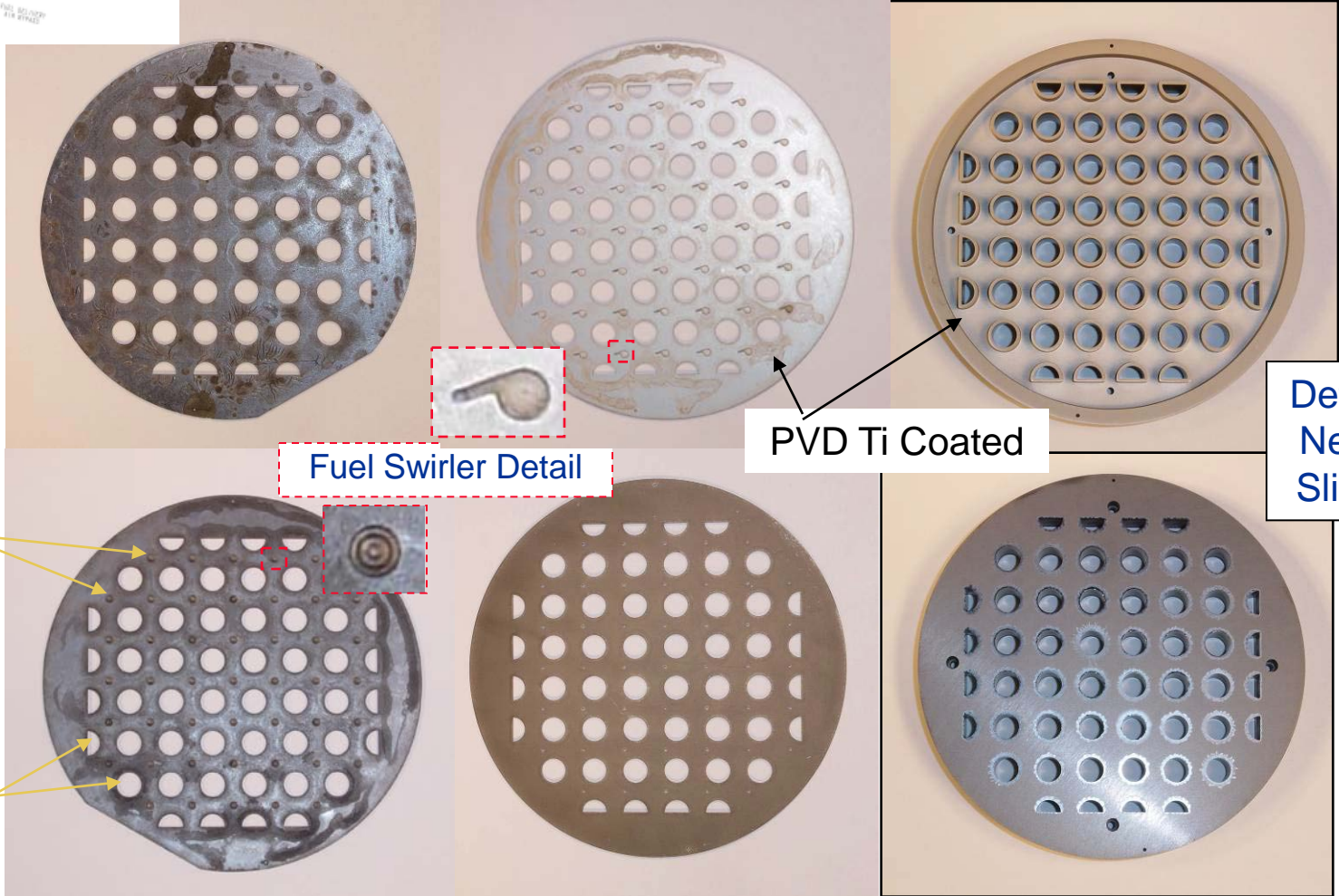
Three Part 10 cm (4") Diameter SiC Injector



Stacking Sequence
Top to Bottom



**Top
Surfaces**



**Small Fuel
Holes**

**Bottom
Surfaces**

**Large Air
Holes**

Fuel Swirler Detail

PVD Ti Coated

Detail
Next
Slide





- Develop lower temperature (less than 1250°C) diffusion bonding conditions with no intermediate phases, no flow, and no micro-cracking.
- Apply diffusion bonding approach to multi-laminate SiC injector.
- Develop SiC to Kovar brazing.

Acknowledgements

- This effort was supported by the NASA Glenn Research Center under the Subsonics and Supersonics Foundation Research Programs.
- The authors would like to thank the following:
 - Dr. Dan L. Bulzan and Robert R. Tacina at NASA GRC for their support and for providing the injector design and requirements.
 - Dr. Robert Okojie of NASA GRC for providing PVD Ti Coated CVD SiC.



Summary and Conclusions

- Diffusion bonds fabricated with the alloyed Ti foil as the interlayer formed five to seven phases in the bond.
 - Microcracks due to the formation of thermally anisotropic phases.
- Diffusion bonds fabricated with the 10 micron PVD Ti coating gave better diffusion bonds than the alloyed Ti foil.
 - Fewer and preferred phases were formed which resulted in bonds without microcracks.
- Processing with thinner interlayers and/or for longer durations gave better diffusion bonds.
- Further optimization is needed to identify lower temperature processing conditions so that phase flow is avoided.