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

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outline

1. Introduction
   - properties and applications of SiC
2. Sample preparations used for diffusion bonding
   - Substrates : SA-Tyrannohex ™ (SA-THX)
   - Interlayers : Ti-Mo foil
3. Experimental results
   - TEM and STEM images of substrates (SA-THX)
   - TEM and STEM images of diffusion bonded samples
4. Discussion about the microstructure of the formed phases by diffusion bonding
   - the orientation relation between the precipitated TiC and Mo-Ti (SS)
5. Summary

SiC is an attractive material

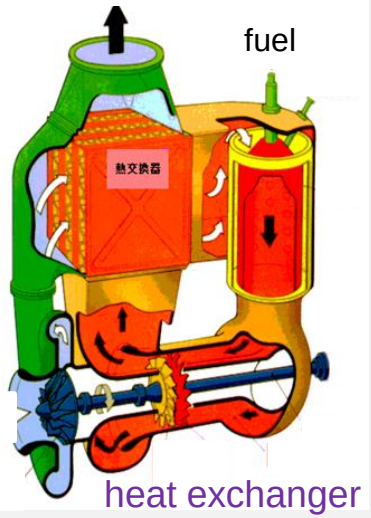
(high-temperature, extreme environment applications)

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

Developed for wide range uses

(not only as a monolithic material, but also in composites)

1. monolithic materials injector applications
2. composites materials combustion liner, nuclear and fusion reactor, turbine engine applications



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

Therefore, new advanced methods are needed.

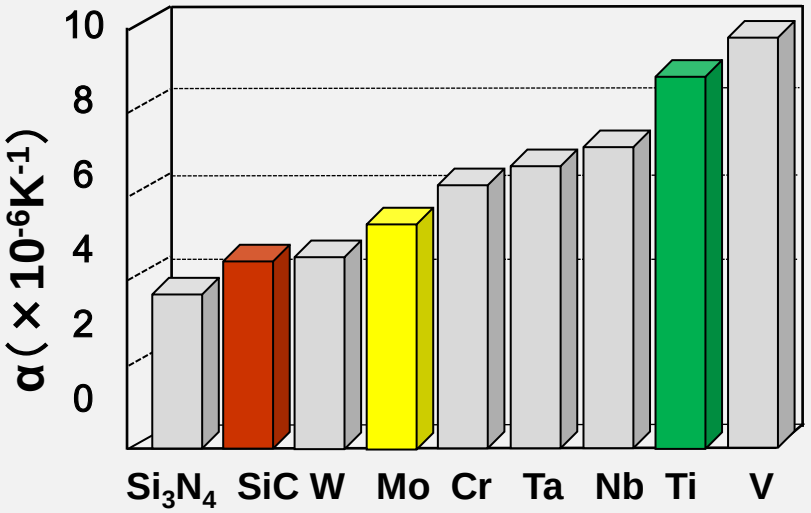
Under those circumstances, one cost-effective solution for fabricating large, complex-shaped components is the joining of simple shaped ceramics.

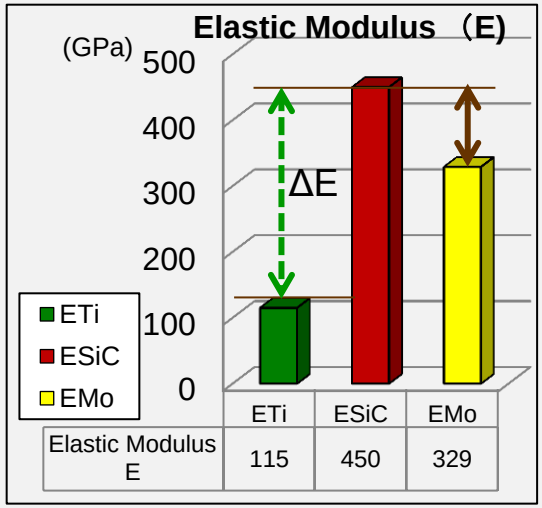
In this study, we are going to focus on

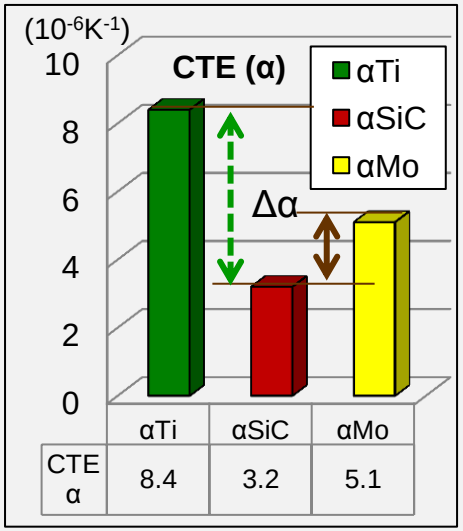
diffusion bonding.

Mismatch of elastic modulus (E) and coefficient of thermal expansion (CTE; α) between substrate and interlayer

We have to pay attention to mismatch of elastic modulus and CTE when we select interlayer material to join SiC.







Both *E* and *CTE* of **Mo** is closer to **SiC** than that of **Ti** .

- **Ti and Mo have been used to join α - SiC.**
- **Better quality bonds formed with Mo than with Ti.**

But,

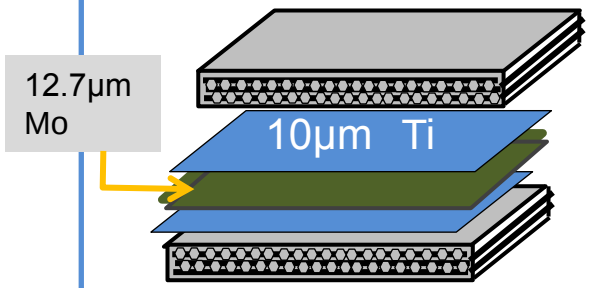
- **Ti can lower the diffusion bonding temperature**.

Therefore, **Ti-Mo** bilayer that possesses both advances of Ti and Mo is also very attractive.

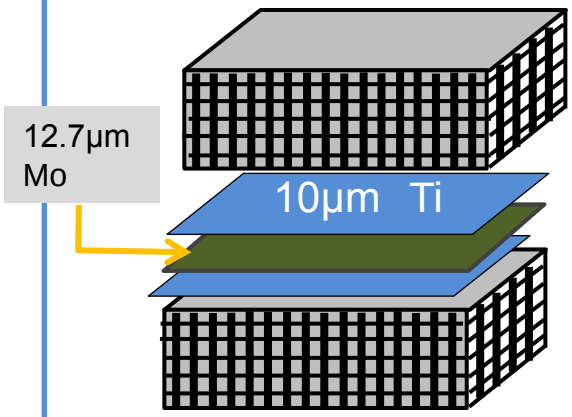
Therefore in this work, we utilize Ti-Mo as interlayers.

Diffusion Bonding of a SA-THX using Ti/Mo metallic Interlayers

Ti-Mo foil



10μm Ti and 12.7 μm Mo interlayer
parallel to SiC fiber



10μm Ti and 12.7μm Mo interlayer
Perpendicular to SiC fiber

Used sample @NASA

SA-THX ...SiC fiber-bonded ceramics, UBE Industries

Ti-foil Mo-foil

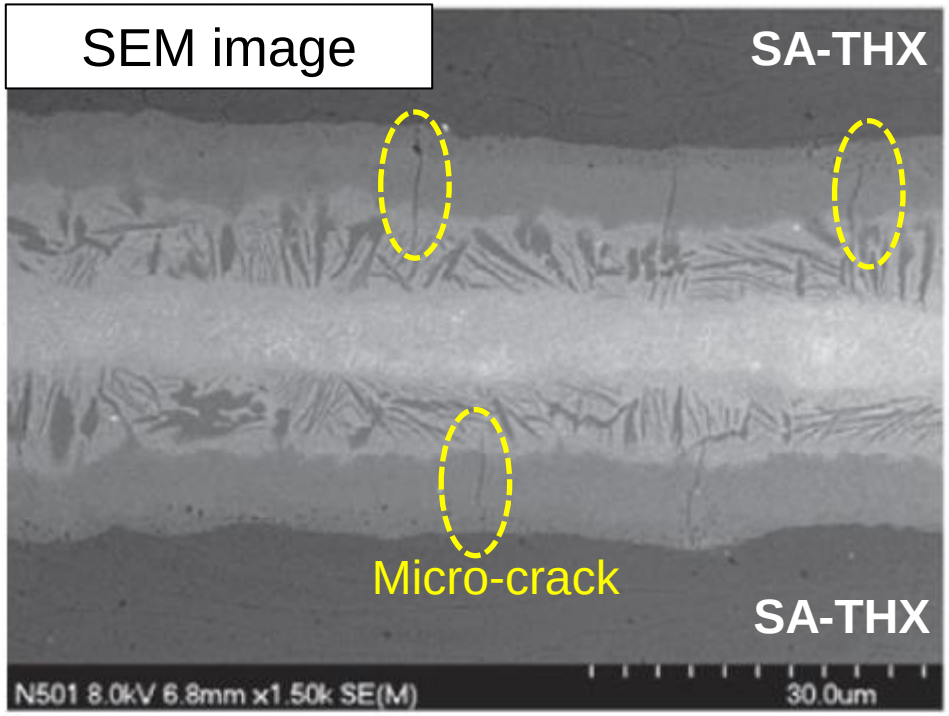
Bonding structure

SA-THX // 10μmTi-12.5μmMo-10μmTi // SA-THX

Bonding process

Hot-press in 1200℃, 4hour, vacuum 30MPa

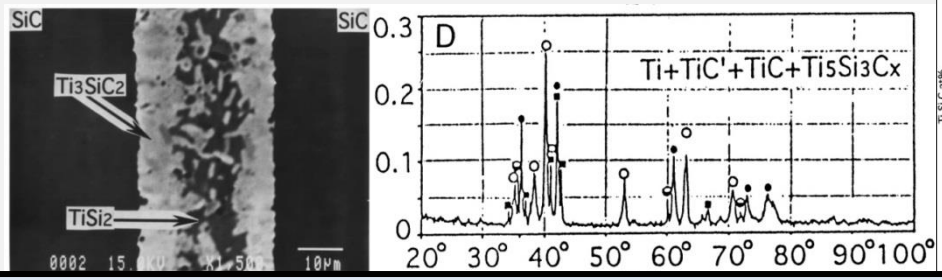
SEM image

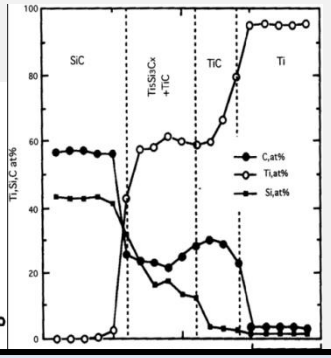


M.C. Halbig, et. al., Ceramics International41(2015)2140–2149

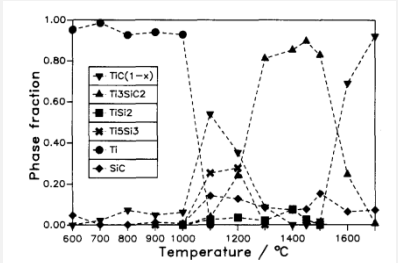
Until now, the phases formed during diffusion bonding have been studied·····.
(to join SiC-SiC using Ti interlayer)

M. Naka et al, Metallurgical and Materials Transactions, A; 28A(1997), 1385-1390



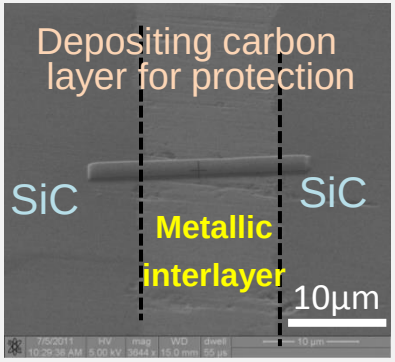


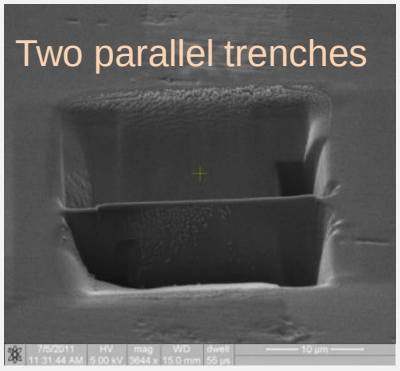
B. Gottselig, et al; J. European Ceramic Society, 6(1990), 153-160

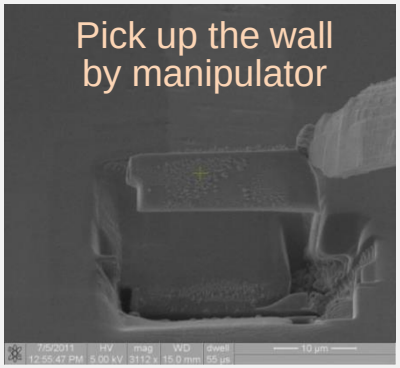


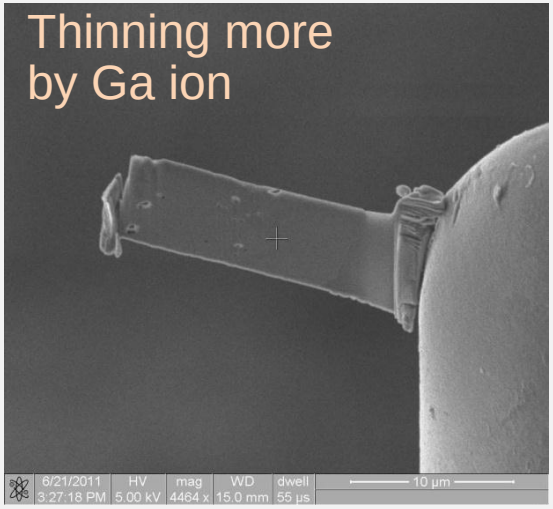
Unfortunately, there has been little literature on TEM observation of the phases formed during diffusion bonding.

Because, it seems very hard to prepare TEM sample from the bonded area. However, recently we successfully obtained a clean, less-damaged, and precisely selected thin specimen from diffusion bonds by using an FIB.









Objectives

We diffusion bonded SiC and SiC (SA-THX and SA-THX) using Ti-Mo 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 bonded SA-THX by TEM and STEM.

2. Characterize the complex microstructure in the diffusion bonded area by TEM observation and SAED analysis.

FIB and Cs-corrected STEM

Focused Ion Beam, FIB
(Hitachi FB-2200)

Cs-corrected STEM
(Hitachi HD-2700)

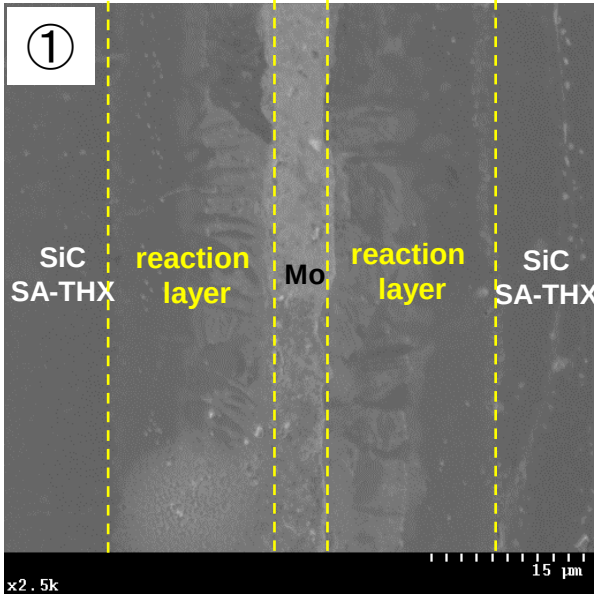
Prepared thin samples for TEM and STEM.

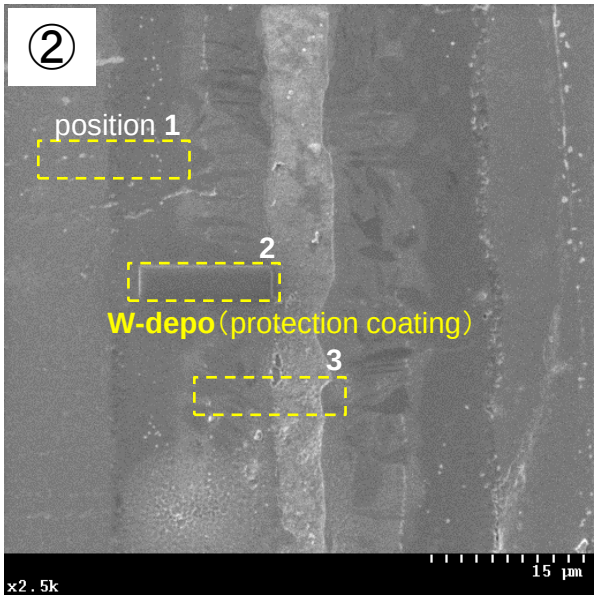
Checked the thin samples prepared by FIB.

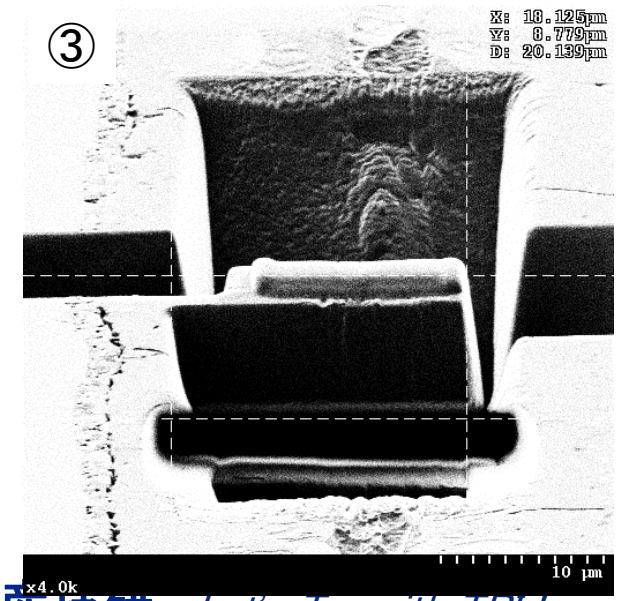
Three-Observation mode:

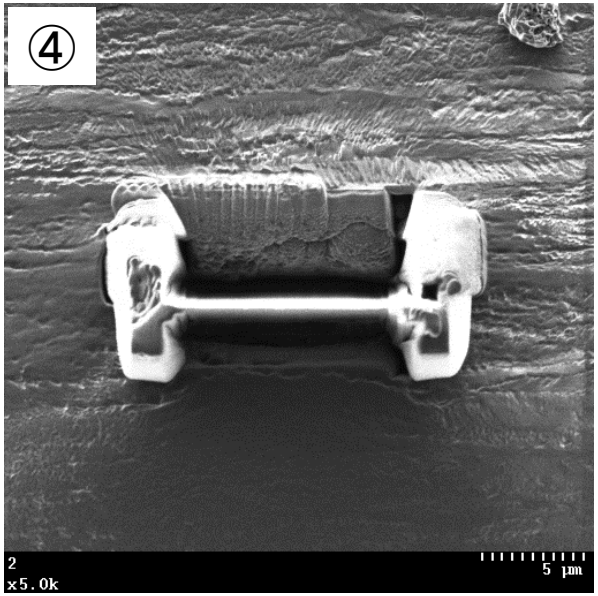
SEM, **BF-STEM** and **HAADF**

Fabricating procedure of the thin sample（SIM image obtained by FIB

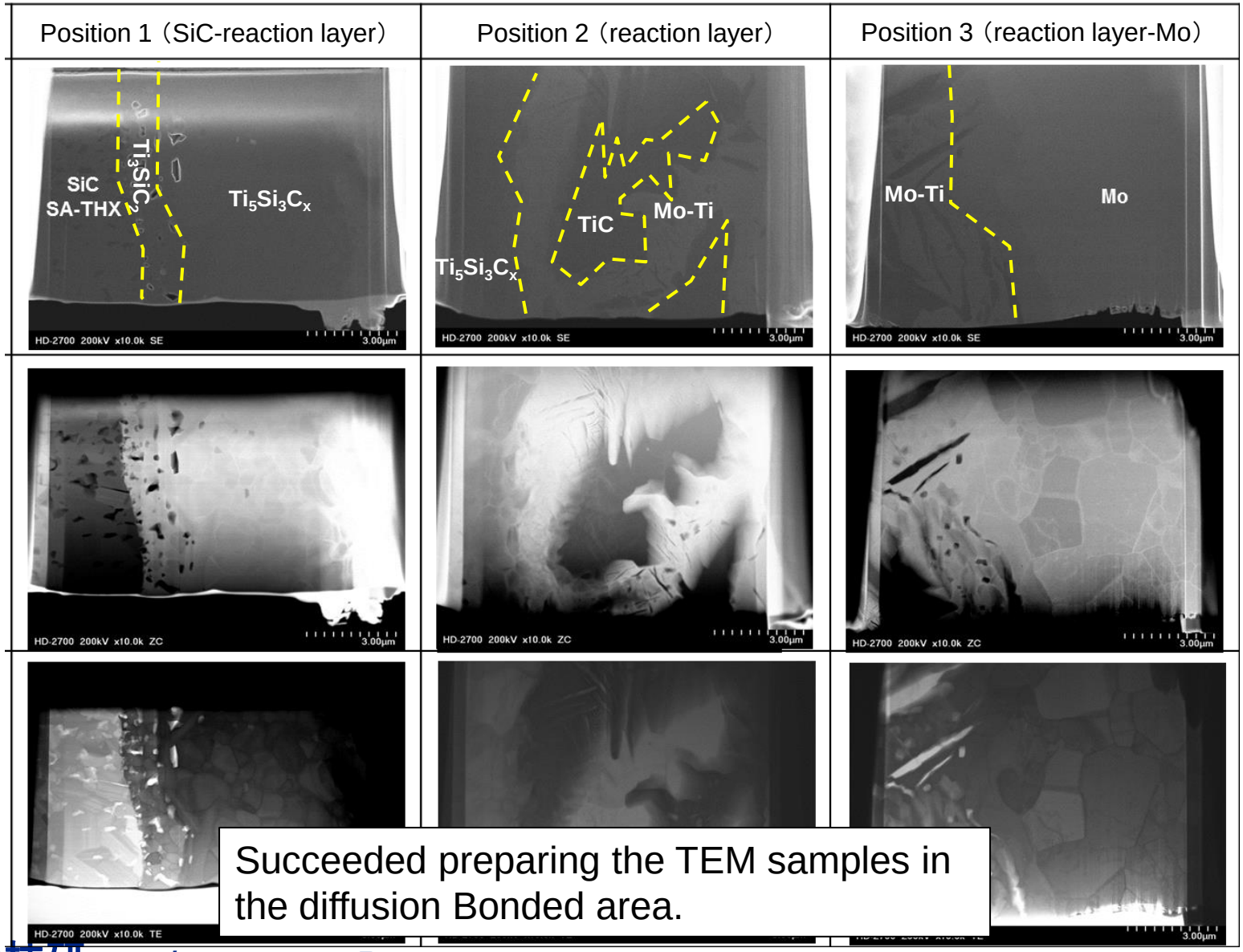








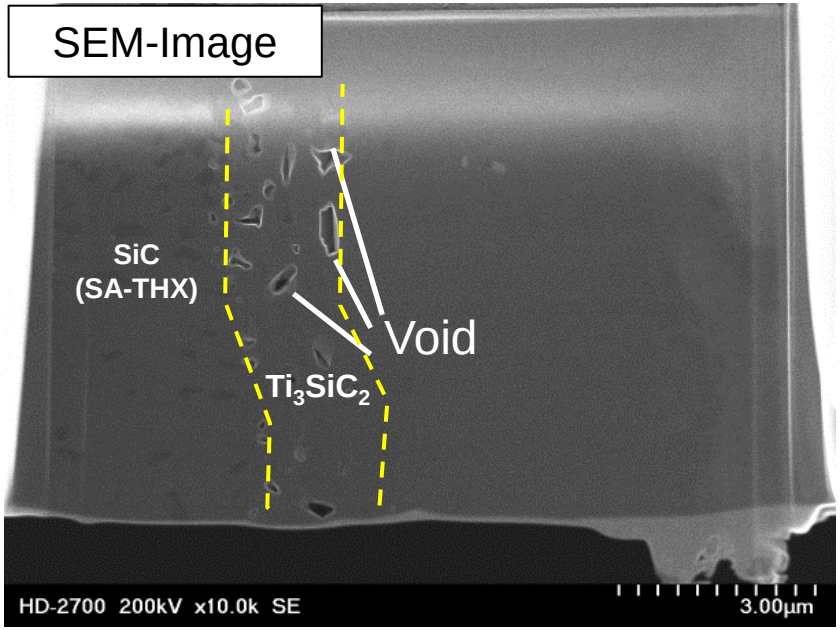
STEM observation of the FIB sample (HD-2700)

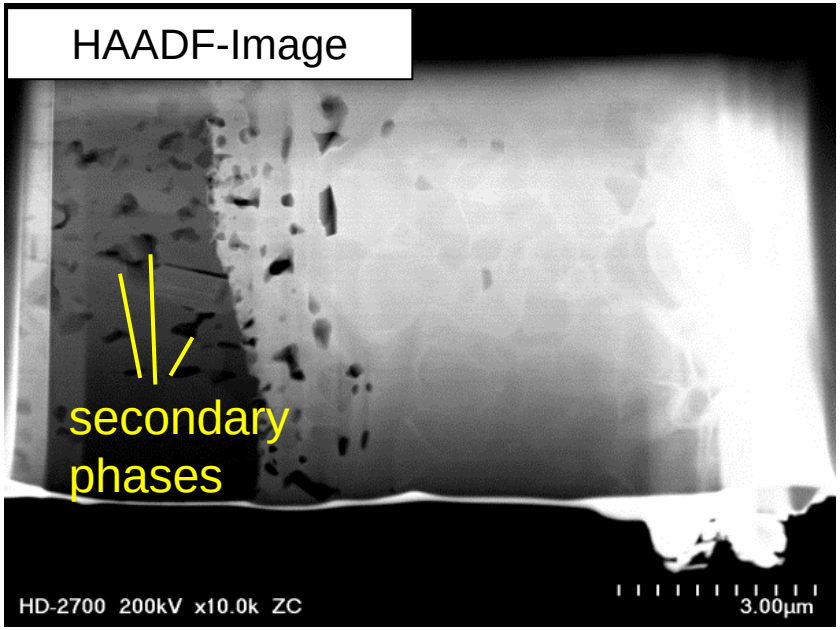


Succeeded preparing the TEM samples in the diffusion Bonded area.

STEM observation of the FIB sample (HD-2700)

Position 1（SiC-reaction layer）

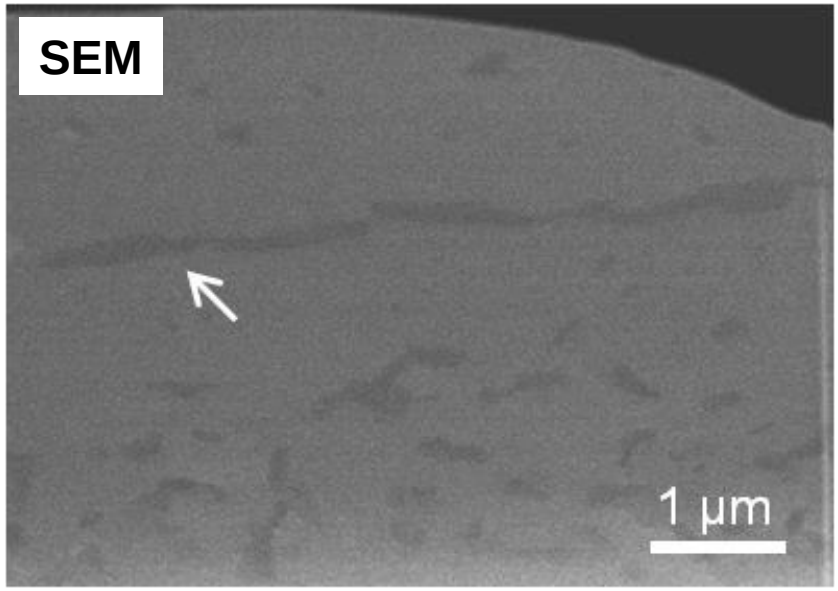


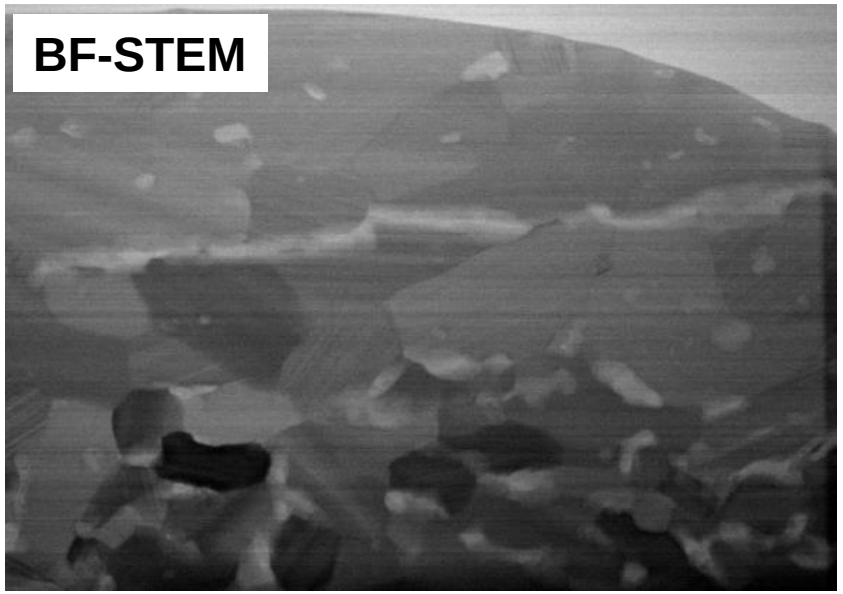


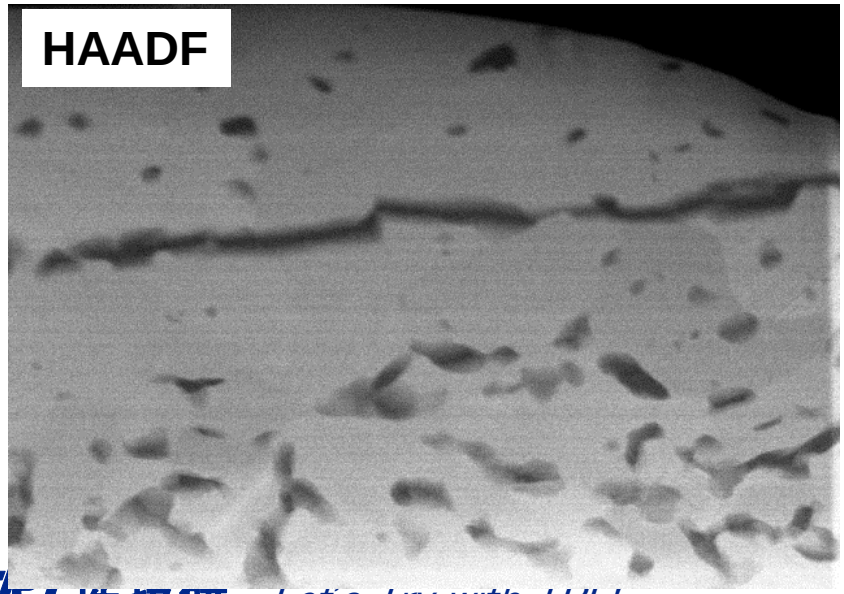
Ti_3SiC_2 phase ---------- Some voids exist.

SA-THX phase --------- Some precipitations (secondary phase) exist.

STEM images (obtained from SA-THX area.)





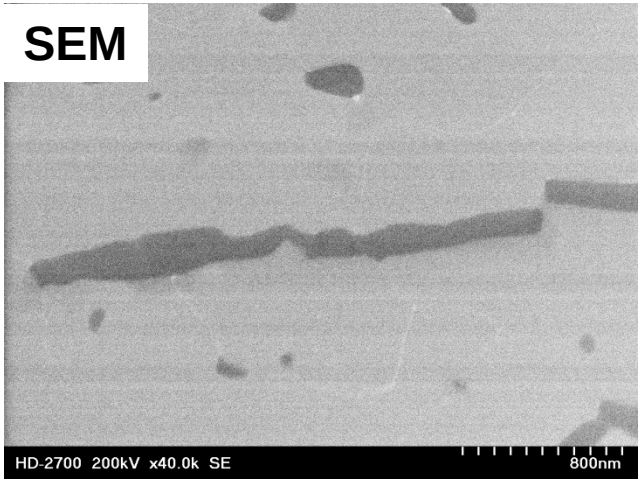


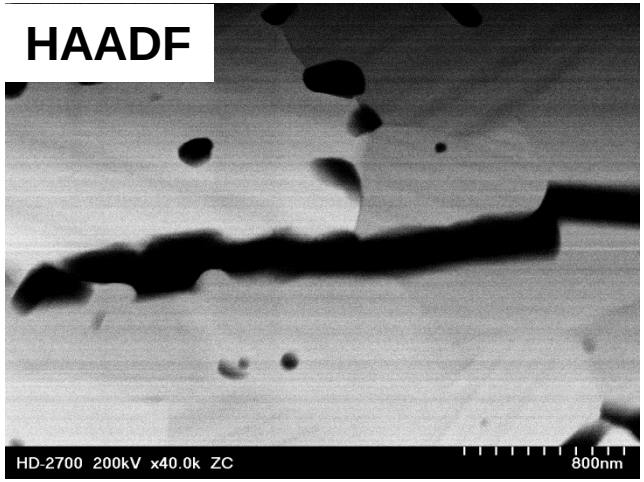
Only in HAADF-image,
the contrast is observed clearly.

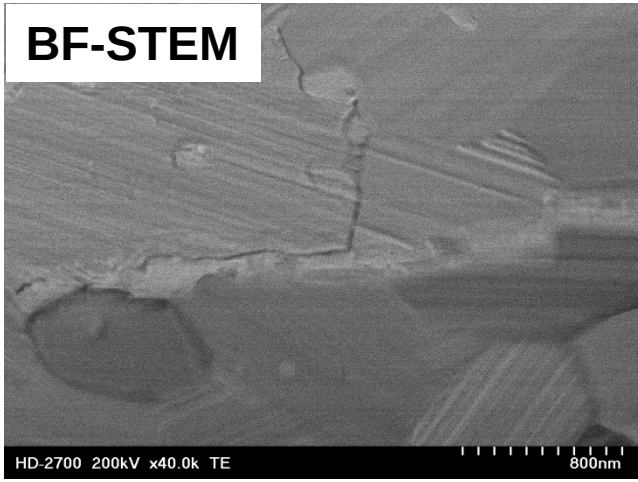
⇒The precipitations is
light element.
(**probably carbon**)

STEM images (obtained from SA-THX area.)

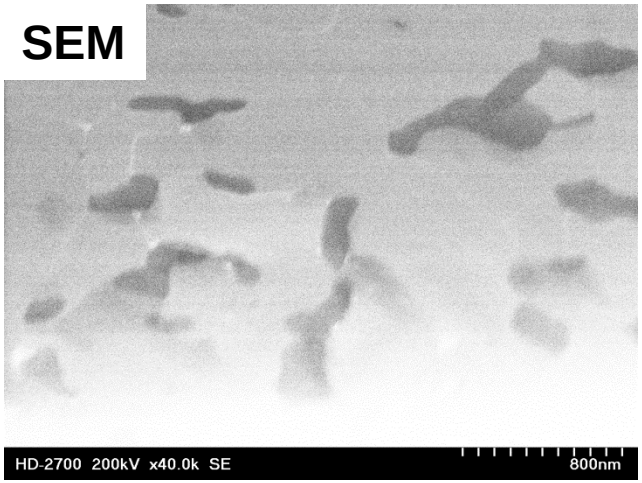
near the boundary of the SA-THX fiber

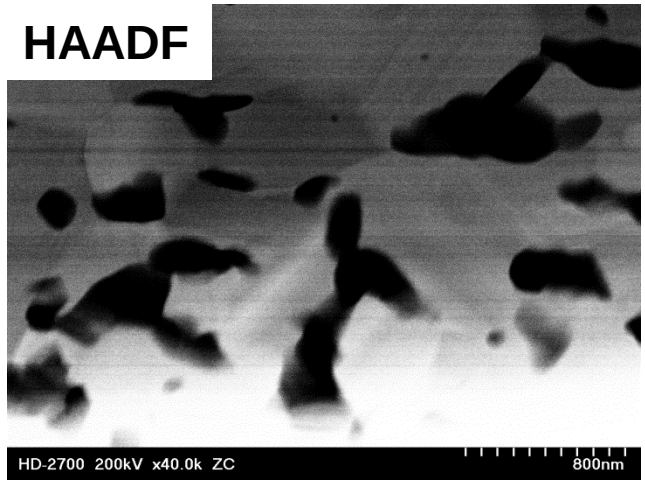


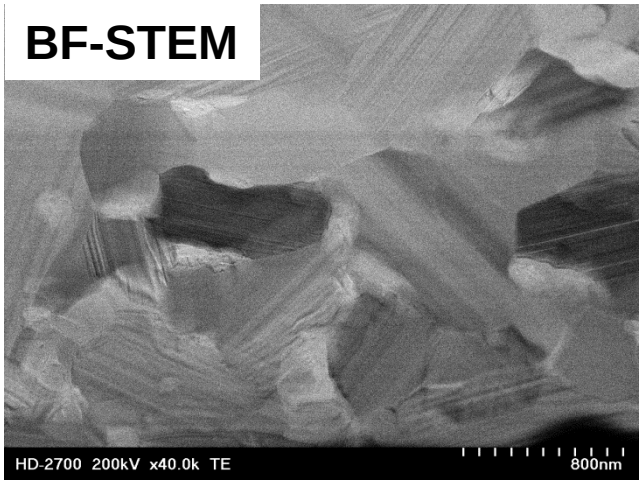




away from the boundary of the SA-THX fiber

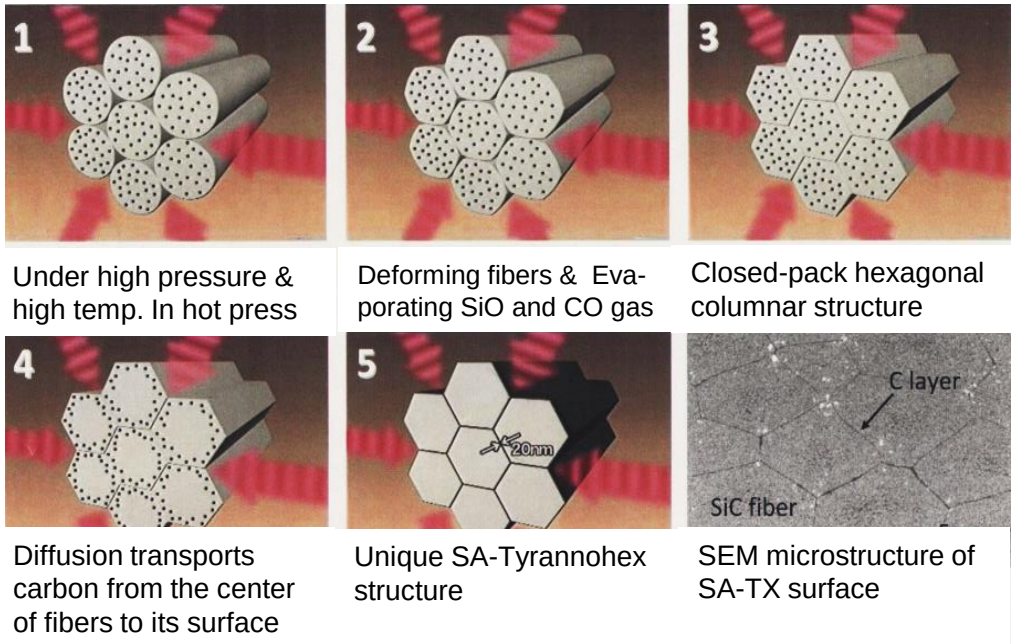






SA-THX forming process

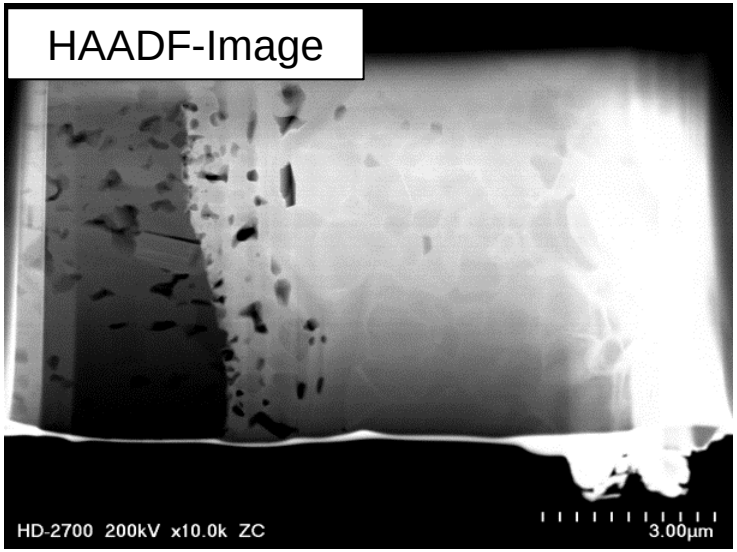
SA-THX forming process



SA-THX has been developed by Dr. T. Ishikawa et al.
T. Ishikawa et al, *Science*, 282, 1295-1297 (1998).
T. Ishikawa et al, *Nature*, 391, 773-775 (1998).
Also, SA-THX is consisting of a highly ordered, closed-packed structure of very fine hexagonal columnar fibers, with a thin interfacial carbon layer between the fibers. The interior of the fiber element was composed of sintered crystalline β-SiC.

⇒ These precipitations stem from residual carbon in SA-THX forming.

Position 1（SiC-reaction layer）



The precipitations are not observed in the reaction layer.

↓

The precipitations don't affect diffusion bonding quality a lot?

Objectives

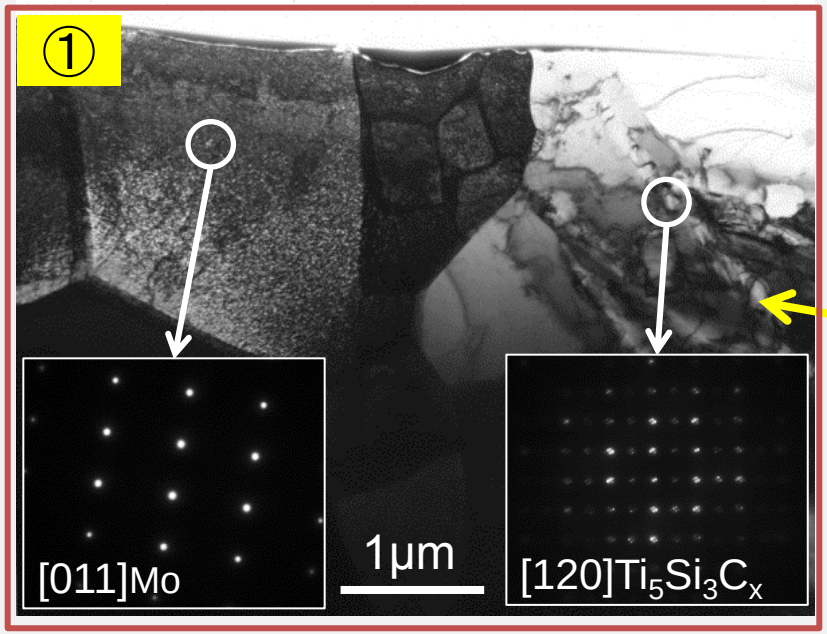
We diffusion bonded SiC and SiC (SA-THX and SA-THX) using Ti-Mo foil metallic interlayer.
We carried out TEM and STEM observations with the diffusion bonded sample prepared by FIB technique.

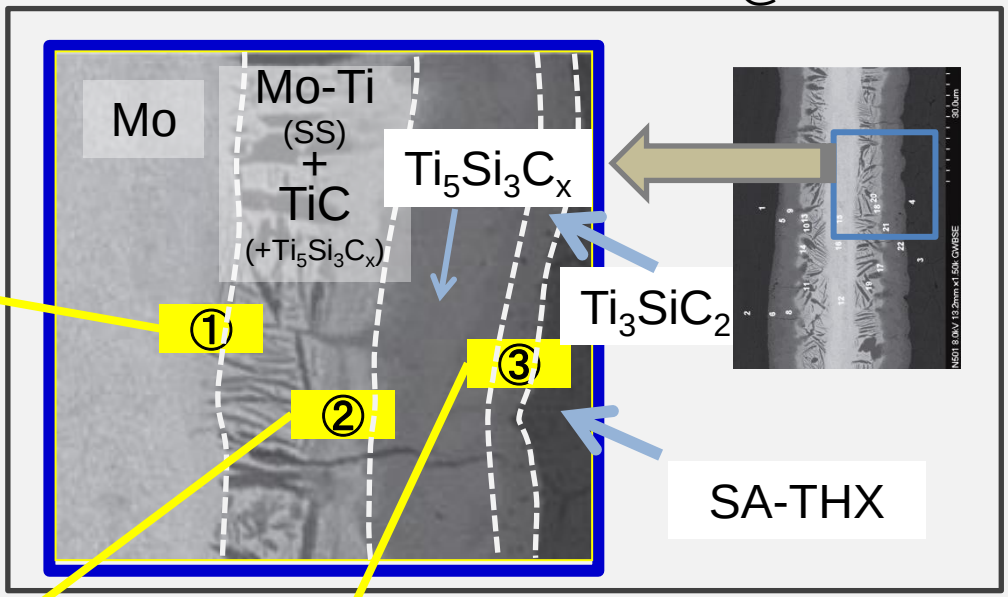
1. Evaluate microstructures of the diffusion bonded SA-THX by TEM and STEM.

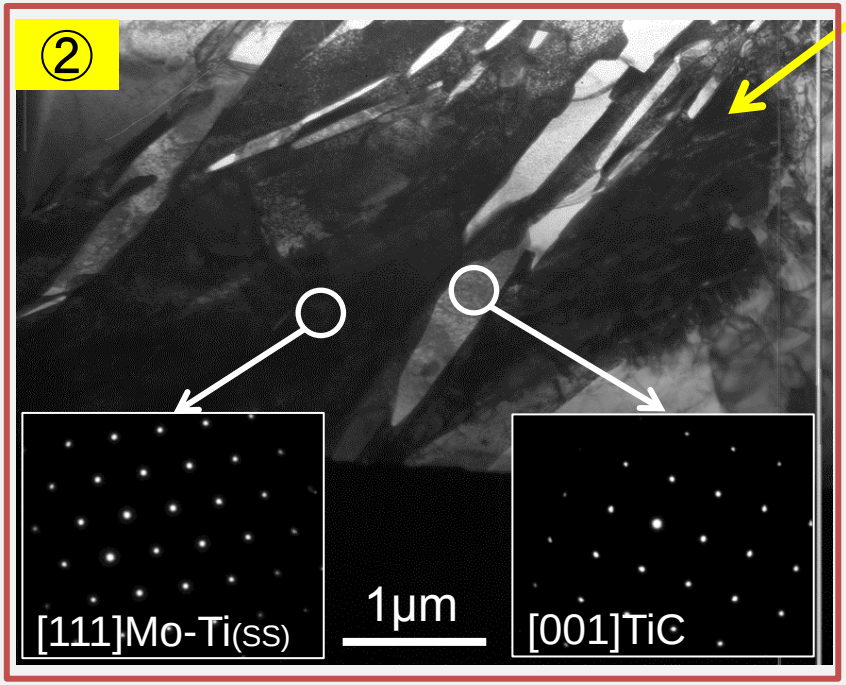
2. Characterize the complex microstructure in the diffusion bonded area by TEM observation and SAED analysis.

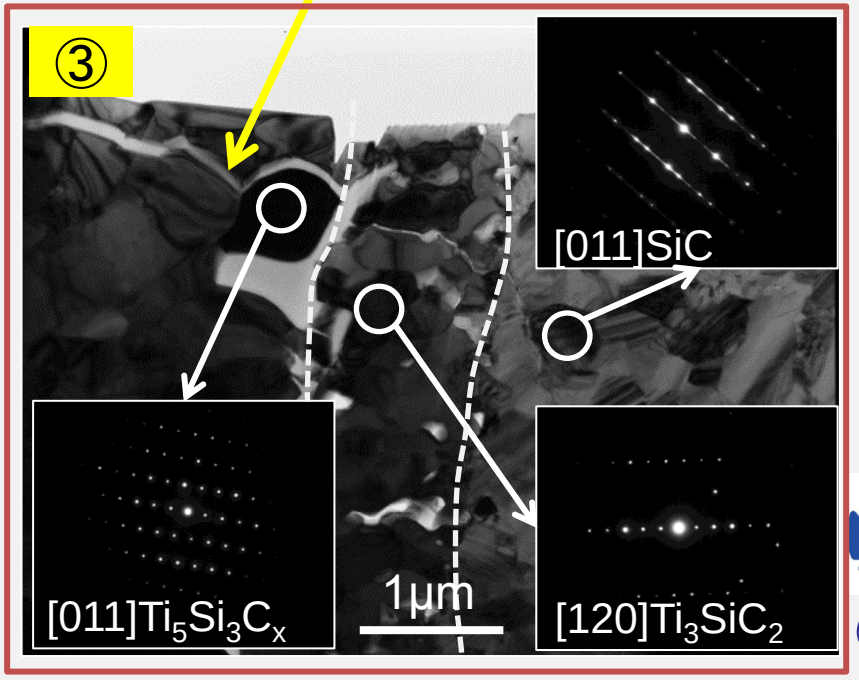
TEM image and SAD patterns of diffusion bond (Ti-Mo foil)

@CMCEE11



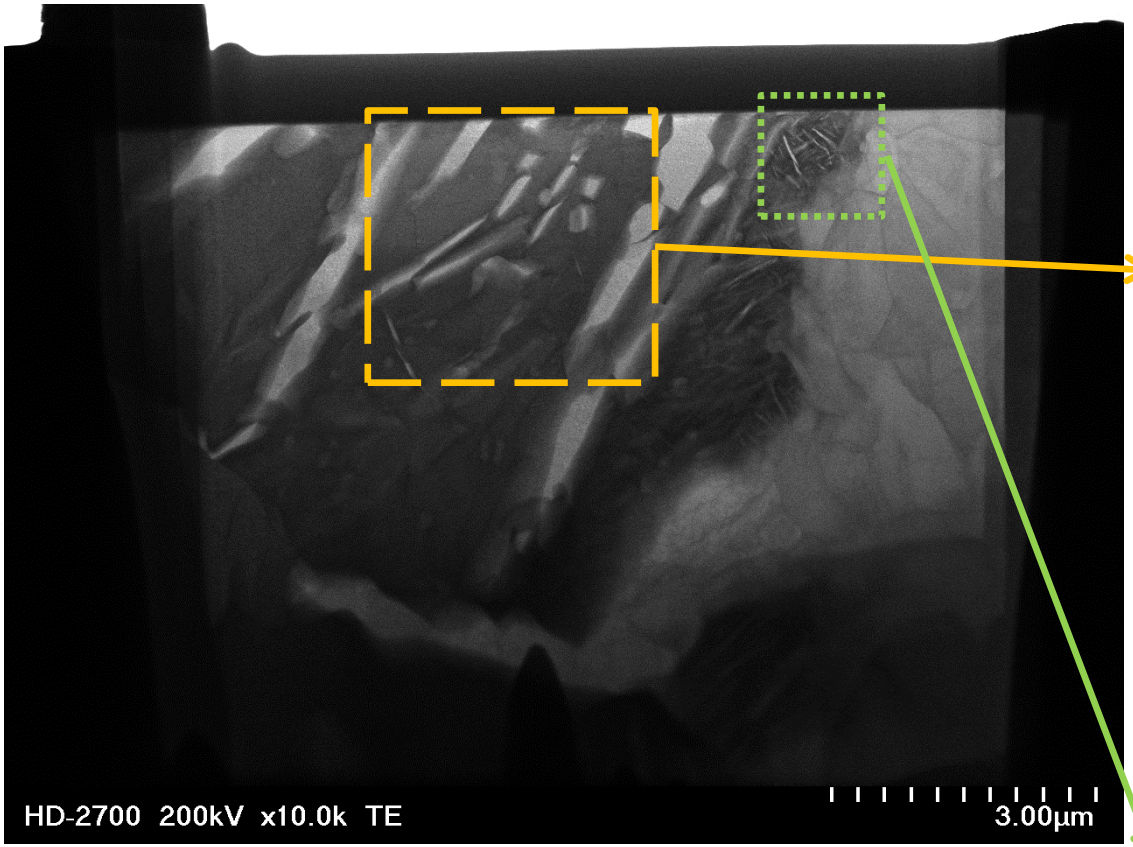


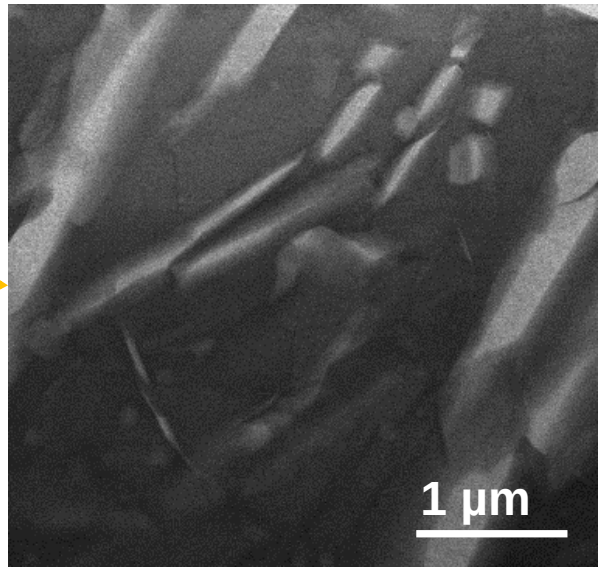


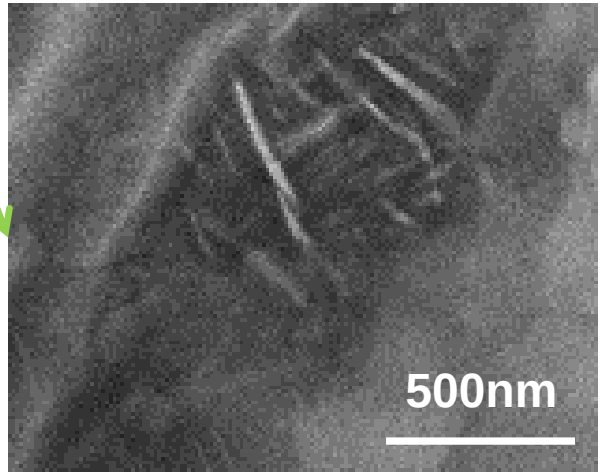


OPU

STEM image of diffusion bond



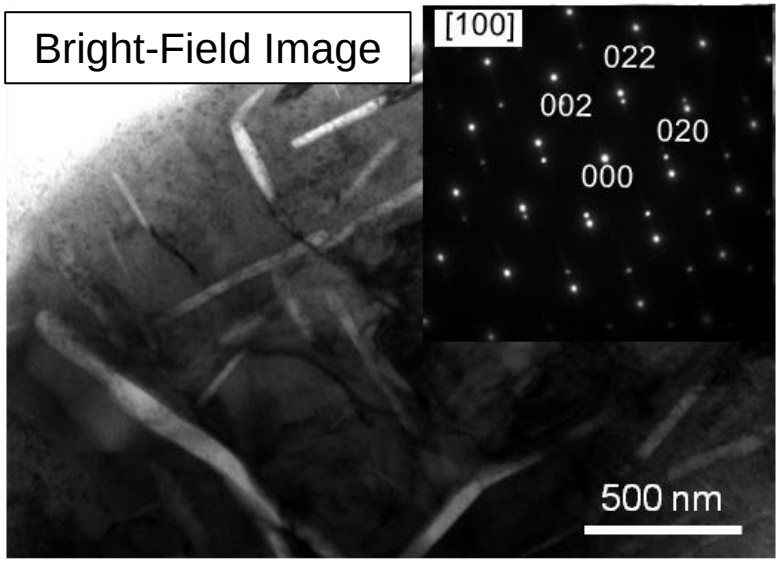




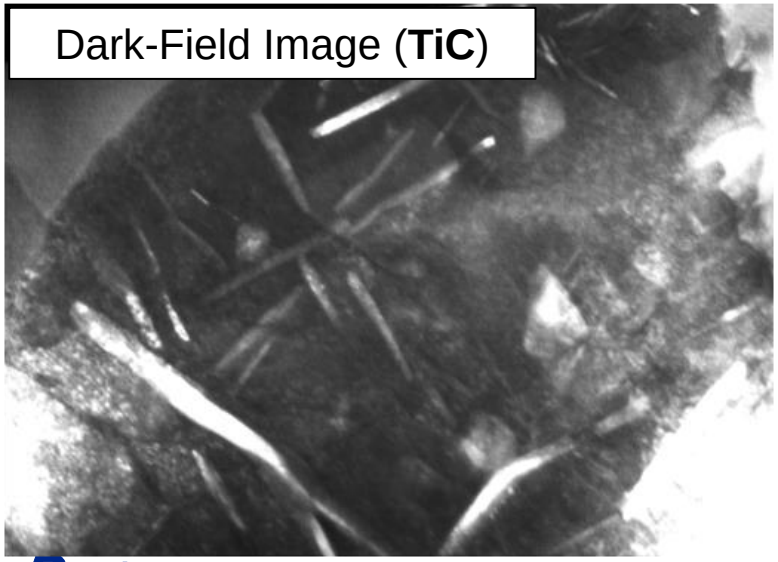
coarse and fine TiC pillars

TEM image and SAED patterns of TiC pillar

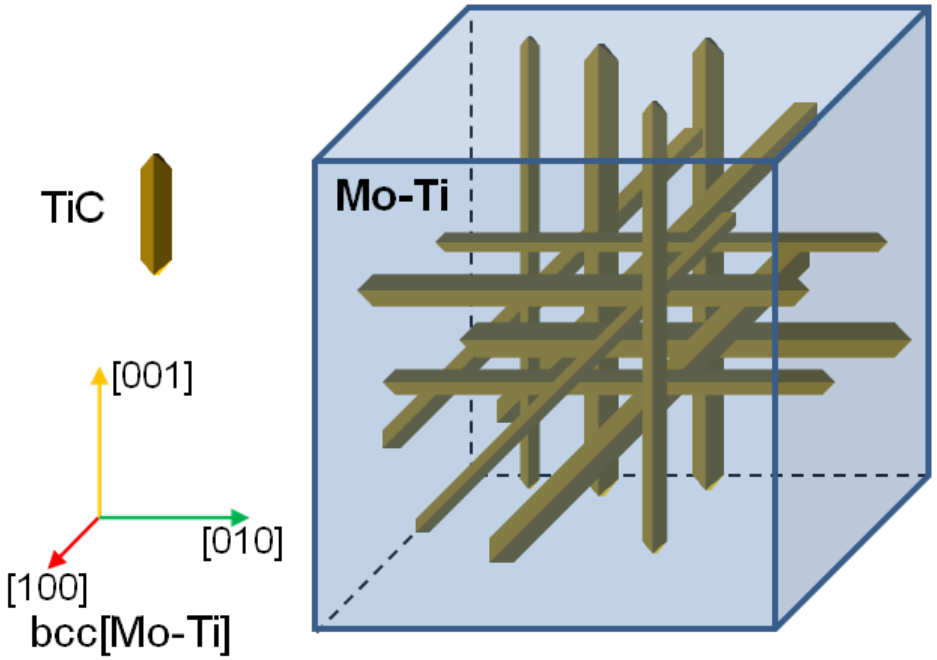
Bright-Field Image



Dark-Field Image (**TiC**)



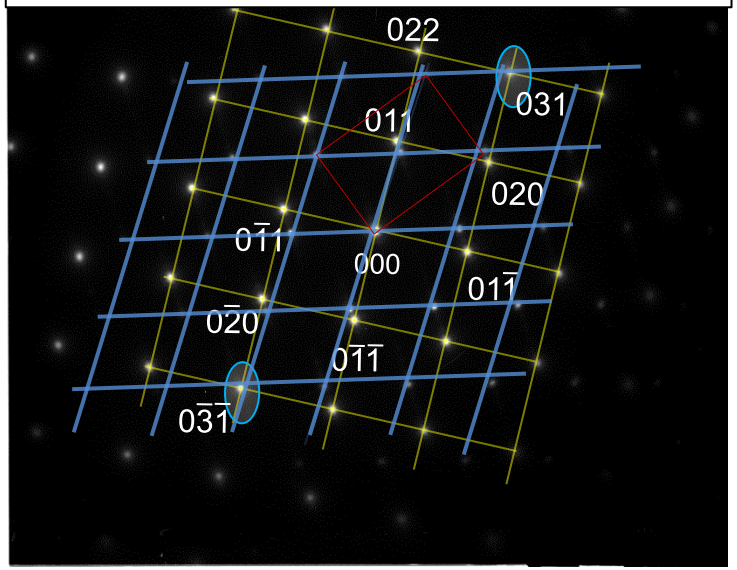
Schematic image of
the location of TiC in [Mo-Ti]ss matrix.



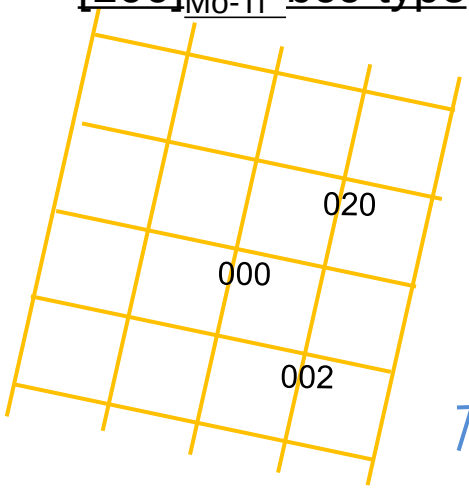
The TiC pillars point to almost <100> direction.

the relation of the crystallographic orientation between Mo-Ti and TiC

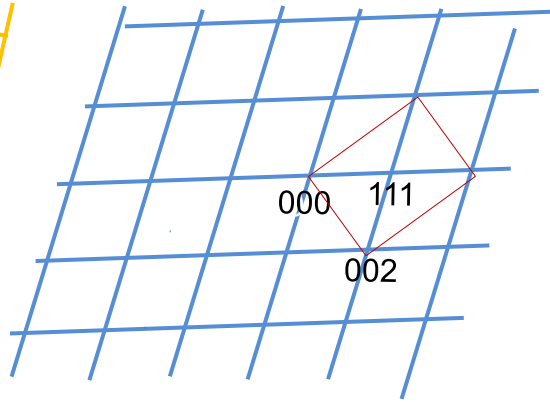
SAED-pattern (Mo-Ti matrix + TiC)



$[100]_{Mo\text{-}Ti}$ bcc-type



$[1\text{-}10]_{TiC}$ NaCl-type



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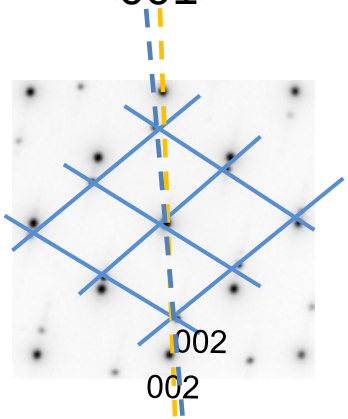
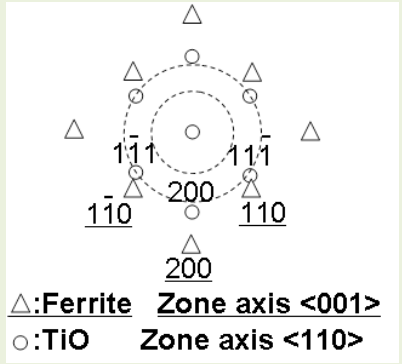


$[100]_{Mo\text{-}Ti}//[1\text{-}10]_{TiC}$

$(002)_{Mo\text{-}Ti}//(002)_{TiC}$



Baker-Nutting's relation

$[100]_{bcc}//[011]_{NaCl}$

$\Rightarrow (002)_{bcc}//(002)_{NaCl}$

R.G. Baker and J. Nutting, Precipitation Process in Steels, I.S.I. Special report,No. 64 (1959).

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. Submicron-sized carbon precipitations were observed in the SA-THX phase away from the boundary of SA-THX fiber. These precipitations did not exist in the reaction phase. It indicates that these precipitations will not affect the diffusion bonding quality a lot.

- 3. TiC pillars were observed around the reaction layer which has a complicated microstructure. The TiC had an orientation relation with the matrix Mo-Ti(SS). In observing from $[100]_{Mo\text{-}Ti}//[011]_{TiC}$ incidence, TiC and Mo-Ti were located in almost $(002)_{Mo\text{-}Ti}//(002)_{TiC}$ relation. It should be considered that precipitated TiC and matrix Mo-Ti has Baker-Nutting's relation that is often seen when NaCl-type material precipitates in a matrix of bcc-type materials.