Title	Joining of SiC Using Amorphous Ti Base Filler Metal(Materials, Metallurgy & Weldability)
Author(s)	Naka, Masaaki; Tanaka, Tasuku; Okamoto, Ikuo
Citation	Transactions of JWRI. 1986, 15(1), p. 49-54
Version Type	VoR
URL	https://doi.org/10.18910/6752
rights	
Note	

Joining of SiC Using Amorphous Ti Base Filler Metal †

Masaaki NAKA*, Tasuku TANAKA*** and Ikuo OKAMOTO**

Abstract

Joining of SiC to SiC or SiC to Cu was made using amorphous $Cu_{50}Ti_{50}$, $Cu_{43}Ti_{57}$ and $Ni_{24.5}Ti_{75.5}$ filler metals, where reaction-sintered SiC containing 13 at%SiC was used.

The joining layer of SiC/SiC joint using Cu-Ti filler is composed of $TiSi_2$, and small amounts of TiSi. The main part of copper in Cu-Ti filler migrates into SiC, and embrittles SiC near the joining layer of SiC/SiC joint. In joining of SiC to Cu using $Cu_{50}Ti_{50}$ filler, large amounts of silicon in SiC migratrate to Cu through the filler and attain to 16 at% Si in Cu, and large amounts of Cu and Ti move into SiC at a large distance from the joint interface. The joining temperature of SiC/SiC joint using Ni-Ti filler at brazing time of 30 min is higher than that of SiC/SiC joint using Cu-Ti filler. The joining layer is composed of $TiSi_2$ and TiSi. The main part of Ni in Ni-Ti filler enters into SiC during brazing of reaction-sintered SiC.

KEY WORDS: (Joining) (Brazing) (Ceramics) (Silicon Carbide) (Copper) (Amorphous Filler) (Amorphous Titanium Alloys) (Titanium Silicide) (Copper Silicide) (Vacuum Brazing)

1. Introduction

Silicon carbide (SiC) in silicon base ceramics that are composed of covalent bonding have attracted recent interests for structural components because of their superior mechanical and corrosion-properties[1]. The physical limitation in producing large ceramics and the worse deformability of ceramics require the joining of ceramics to ceramics or ceramics to metals in practical application. In this paper the joining of reaction-sintered SiC to SiC using Cu-Ti and Ni-Ti amorphous filler, and the joining of reaction-sintered SiC to copper using Cu-Ti amorphous filler are made, where the amorphous filler metals provide the simplicity and reliability to the joining method[2,3]. The joining mechanism is further made clear.

2. Experimentals

The reaction-sintered SiC used in joining contains 13 wt% Si and 1 wt% SiO_2. The characteristic properties of amorphous Ti base fillers of $Cu_{50}Ti_{50}$, $Cu_{50}Ti_{57}$ and $Ni_{24.5}Ti_{75.5}$ used are shown in **Table 1**. The amorphous fillers 1 cm wide and 45 μm thick were produced by ejecting the liquid alloys onto the outer surface of a highly rotating wheel[3]. SiC of 15 mm in diameter and 3 mm in thickness, and SiC or Cu of 6 mm in diameter and 3 mm in thickness were used for a lap joint. The joining was conducted in 1×10^{-5} torr or above at desired joining temperature and time. The heating rate up to brazing temperature without loading was 20°C/min, and the cooling rate after brazing was 20°C/min down to 600°C, and then 1°C/min to room temperature. The joining strength of the lap joint was determined by shear fracture loading using a cross head speed of 1 mm/min at room temperature. The microstructures and element distribution of specimens joints were determined by means of scanning electron microscope, EDX and EPMA microanalyser, and X-ray diffractometer, respectively.

Table 1 Physical properties of amorphous Ti base filler metals

	Nominal composition (at%)			Liquidus temperature (°C)	Thickness (μm)
	Ti	Cu	Ni		
$Cu_{50}Ti_{50}$	50	50	-	975	50
$Cu_{43}Ti_{57}$	57	43	-	955	45
$Ni_{24.5}Ti_{75.5}$	75.5	-	24.5	955	45

3. Results and Discussion

3.1 Joining of SiC/SiC joint with Cu-Ti filler

3.1.1 Joining strength of SiC/SiC joint

Figure 1 shows the brazing temperature dependence of joining strength of SiC/SiC joint using $Cu_{50}Ti_{50}$ and $Cu_{43}Ti_{57}$ fillers at constant brazing time of 1 min. The joining strength of the joint with $Cu_{43}Ti_{57}$ filler is higher than that with $Cu_{50}Ti_{50}$ filler. For instance, the maximum strength of the joint brazed at 1025°C for 1 min with $Cu_{50}Ti_{50}$ is 3.8 kgf/mm^2, and the maximum strength of the joint brazed at 1000°C for 1 min with $Cu_{43}Ti_{57}$ is 8 kgf/mm^2. As described in observing microstructures of the joints, the joining with filler containing higher

† Received on April 25, 1986
* Associate Professor
** Professor
*** Graduate Student (Present address, Hitachi Co. Ltd.)

Transactions of JWRI is published by Welding Research Institute of Osaka University, Ibaraki, Osaka 567, Japan

titanium content gives the less degree of embrittlement near the joining layer in SiC. This gives rise to the higher joining strength of SiC/SiC joint. The change of joining strength of SiC/SiC joint with $Cu_{50}Ti_{50}$ filler with brazing time at joining temperature of 1000°C and 1025°C is shown in **Fig. 2**. At joining temperature of 1000°C the joining strength of the joint decreases with an increase in joining time through the maximum of 6.5 kgf/mm^2, and at joining temperature of 1025°C the strength of the joint decreases with an increase in joining time from 3.3 kgf/mm^2 at joining time of 1 min. The joining strength of SiC/SiC joint is higher as the joining temperature is lower. As described below in observing microstructures of the joint, the reaction between the metal in the filler and free-silicon in SiC is suppressed by decreasing the joining temperature.

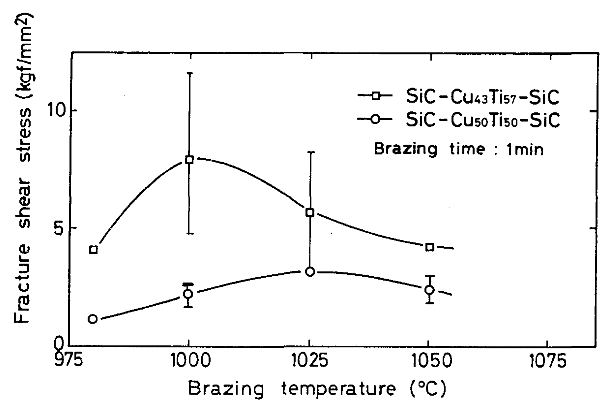

Fig. 1 Change in shear strength of SiC/SiC joint using Cu-Ti filler with brazing temperature at brazing time of 1 min.

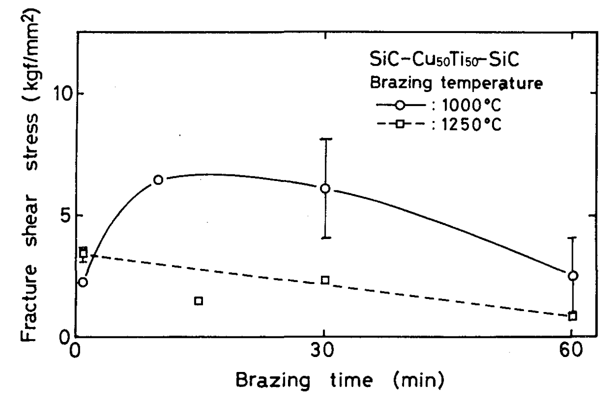

Fig. 2 Change in shear strength of SiC/SiC joint uusing $Cu_{50}Ti_{50}$ filler with brazing time at brazing temperature of 1000° and 1025°C.

3.1.2 Microstructures of SiC/SiC joint

Figure 3 shows the microstructure of SiC/SiC joint brazed at 1100°C for 30 min using $Cu_{50}Ti_{50}$ filler. The structure-changed layer is observed on both side of brazed layer. The brittle phases formed by the reaction between metals in the filler and free-silicon in SiC are partly lacked in cutting and polishing the joint. **Figure 4** shows SEM microstructures and line analyses for Ti, Si and Cu of the same joint in Fig. 3, and spot analyses at the brazed layer. The brazed layer is mainly composed of Ti and Si, and a small amount of Cu. The main part of copper in the filler moves into SiC. The line analyses of the brazed layer shows that the brazed layer is composed of silicides though the spot analyses of the joint in Fig. 4 and the standard sample indicate that the brazed layer at 1100°C

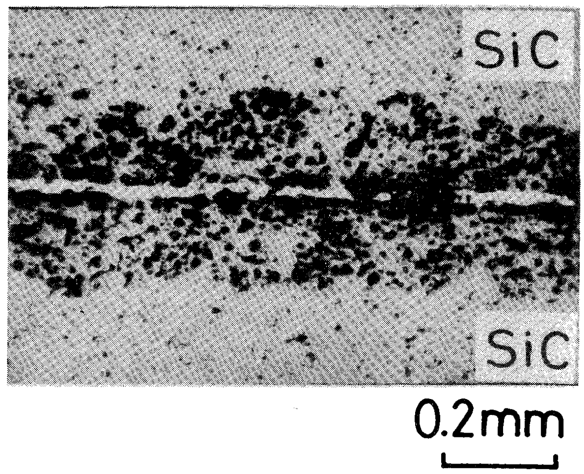

Fig. 3 Micrograph of joining layer and embrittled region of SiC/SiC joint using $Cu_{50}Ti_{50}$ filler brazed at 1100°C for 30 min.

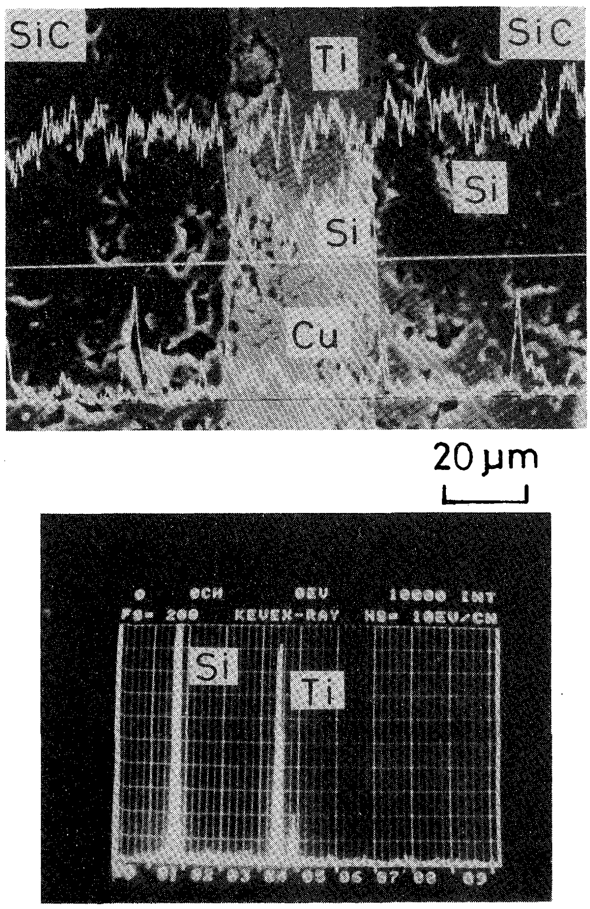

Fig. 4 Microstructure and spot analyses of brazed layer in SiC/SiC joint using $Cu_{50}Ti_{50}$ filler brazed at 1100°C for 30 min.

for 30 min is mainly composed of $TiSi_2$. The spot analyses of embrittled SiC at 0.2 mm from the brazed layer are shown in **Fig. 5**. Ti and Cu in the filler move into SiC independently in (i) and (ii) or together in (iii). The main part of titanium in the filler remaines in the brazed layer and the large amounts of copper in the filler migrate into the reaction-sintered SiC.

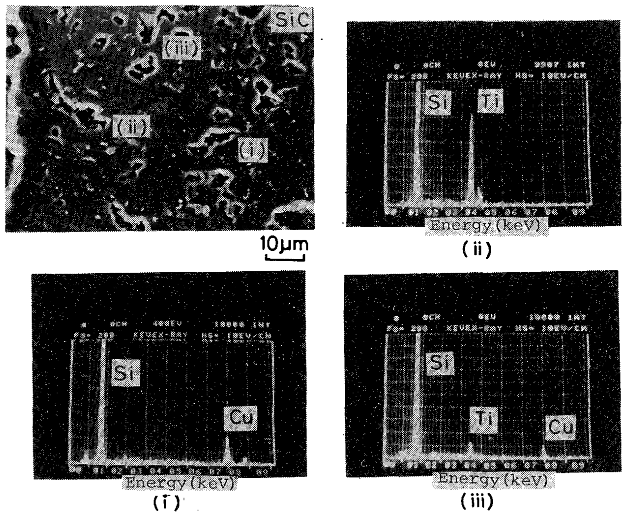

Fig. 5 Microstructure and spot analyses in embrittled SiC part of SiC/SiC joint using $Cu_{50}Ti_{50}$ filler brazed at 1100°C for 30 min.

Figure 6 shows the diffraction pattern of the fracture surface of SiC/SiC joint brazed at 1100°C for 30 min. $TiSi_2$ shown in Fig. 2 and a small amount of TiSi and ϵ-$Cu_{15}Si_4$ are observed in the diffraction pattern. **Figure 7** demonstrates the EPMA line analyses of C, Si and Ti of SiC/SiC joint brazed at 1000°C for 30 min. No EPMA peak of carbon and the low carbon content in the brazed layer indicate that the titanium carbide such as TiC is not formed. The uniform line analysis of Ti in the brazed layer represents that the amount of TiSi in the diffraction pattern in Fig. 6 is small.

3.2 Joining of SiC/Cu joint with Cu-Ti filler

3.2.1 Joining strength of SiC/Cu joint

Figure 8 shows the joining temperature dependence of joining strength of SiC/Cu joint brazed at the constant brazing time of 30 min using $Cu_{50}Ti_{50}$ filler. The strength decreases with an increase in brazing temperature. Since the copper in the joint is melted at brazing temperatures of 1025 and 1050°C, the strength is estimated from the fracture area after fracture shear testing. The formation of Cu-Si phases that possess low melting points are formed by moving of Si from SiC to Cu in the joints through the brazed layer. This leads to melt the copper of the joint at the brazing temperature below 1083°C of melting point of copper.

3.2.2 Microstructures of SiC/Cu joint

Figure 9 shows the change of microstructure of SiC/Cu joint brazed at 1025°C with brazing time using $Cu_{50}Ti_{50}$ filler. The thickness of black region in SiC near the brazed layer changes from 0.02 mm at brazing time of 5 min (a) to 0.1 min at brazing time of 30 min (b). The black region at brazing time of 60 min does not change the thickness, but increases the metallic color. The line

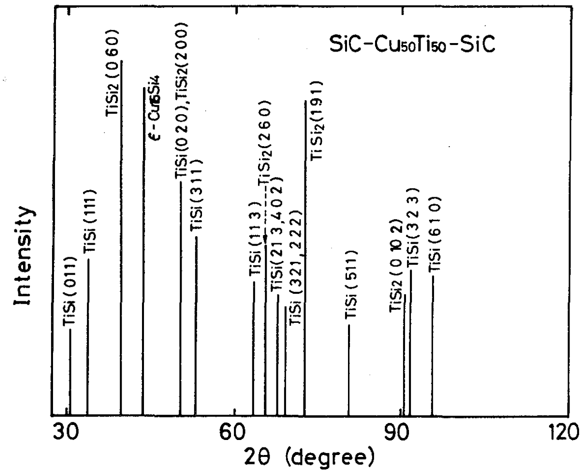

Fig. 6 X-ray diffraction pattern of fracture surface using $Cu_{50}Ti_{50}$ filler brazed at 1100°C for 30 min.

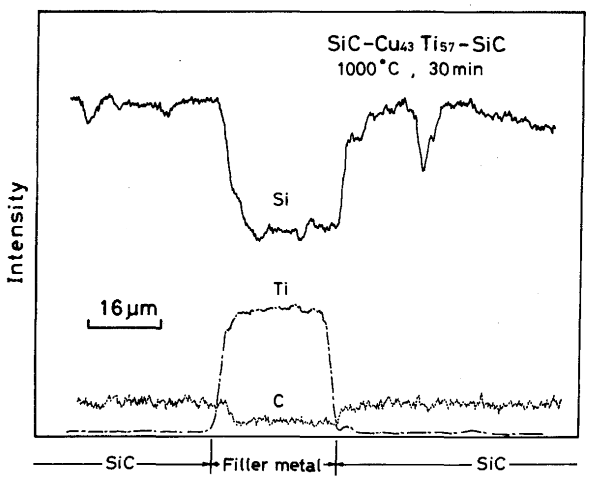

Fig. 7 Electron microprobe analyses for Ti, Si and C in SiC/SiC joint using $Cu_{43}Ti_{57}$ filler brazed at 1000°C for 30 min.

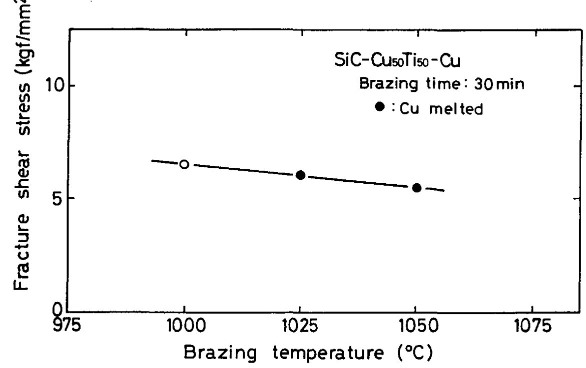

Fig. 8 Change in shear strength of SiC/Cu joint using $Cu_{50}Ti_{50}$ filler with brazing temperature at brazing time of 30 min.

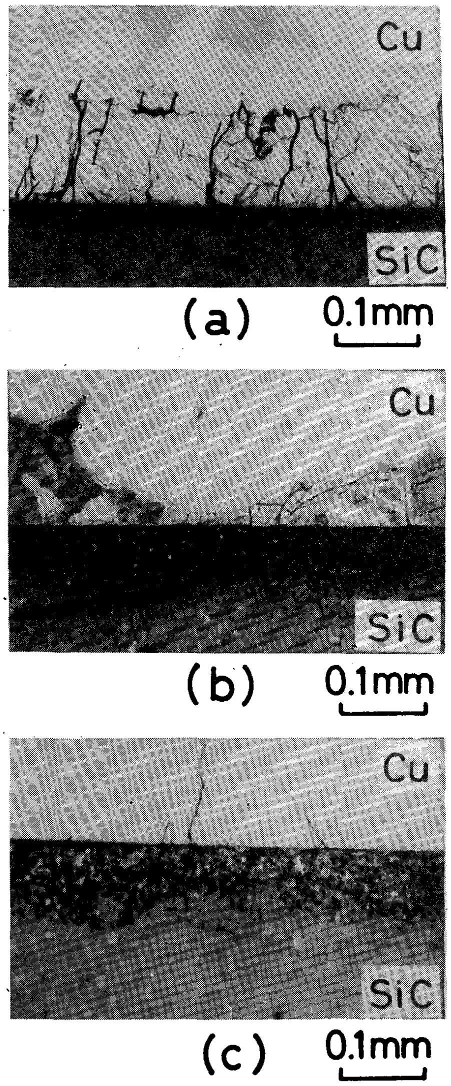

Fig. 9 Microstructures at joining interface of SiC/Cu joint using $Cu_{50}Ti_{50}$ filler brazed at 1025°C for 5 min (a), 30 min (b) and 60 min (c).

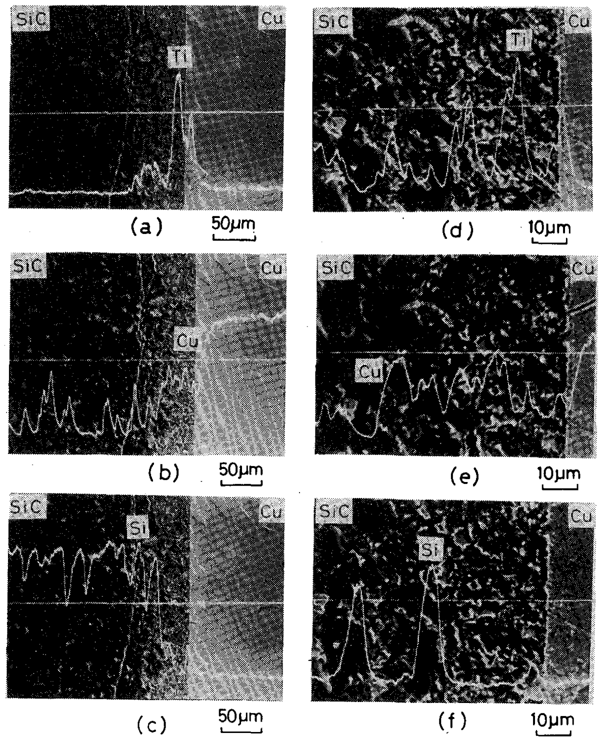

Fig. 10 Microstructures and line analyses for Ti, Cu and Si in SiC at joining interface of SiC/Cu joint using $Cu_{50}Ti_{50}$ filler brazed at 1025°C for 30 min.

analyses of Ti, Cu and Si of SiC/Cu joint brazed at 1025°C for 30 min in **Fig. 10** reveal that the large amounts of Cu and Ti migrate into the black region in Fig. 9. The line analyses of Ti, Cu and Si in **Fig. 11** indicates that copper preferentially moves into the black region. This migration of copper into SiC is caused by the reaction between Cu and free-silicon in SiC. The large amounts of silicon migrate through the brazed layer from SiC to Cu in SiC/Cu joint brazed at 1025°C for 30 min. The analyses of silicon in the joint and the standard sample indicate the content silicon attains to 16 at% Si in Cu in the joint. The whole of copper material transforms to γ-Cu_5Si (m.p. 830°C). This corresponds to the melting of copper material during joining.

3.3 Joining of SiC/SiC joint with Ni-Ti filler

3.3.1 Joining strength of SiC/SiC joint

Figure 12 represents the brazing temperature dependence of joining strength of SiC/SiC joint brazed at a constant brazing time of 30 min using $Ni_{24.5}Ti_{75.5}$ filler. The joining begins at temperatures above 1150°C. The maximum strength of 5 kgf/mm^2 obtained at 1025°C using $Ni_{24.5}Ti_{75.5}$ filler is lower than the maximum strength of 8 kgf/mm^2 of SiC/SiC joint brazed at 1000°C for 1 min using $Cu_{43}Ti_{57}$ filler. The brazing temperature in SiC/SiC joint using Ni-Ti filler is higher than that using Cu-Ti filler. **Figure 13** shows the brazing time dependence of joining strength of SiC/SiC joint at a constant brazing temperature of 1025°C. The joining strength through the maximum of 6 kgf/mm^2 decreases with increasing brazing time.

3.3.2 Microstructures of SiC/SiC joint

Figure 14 shows the microstructure and line analyses of Ti, Ni and Si of SiC/SiC joint brazed at 1250°C for 30 min using $Ni_{24.5}Ti_{75.5}$ filler. Though the nickel content in the brazed layer using Ni-Ti filler is higher than the copper content in the brazed layer using Cu-Ti filler, the large amounts of nickel in the filler migrate into reaction-sintered SiC, and the brazed layer is extremely embrittled, and is lacked during cutting and polishing the joint. **Figure 15** shows the EPMA line analyses of C, Ti and Si corresponds to $TiSi_2$ and TiSi that are identified by X-ray diffraction analysis in **Fig. 16**. The diffraction lines of SiC are eliminated in the figure.

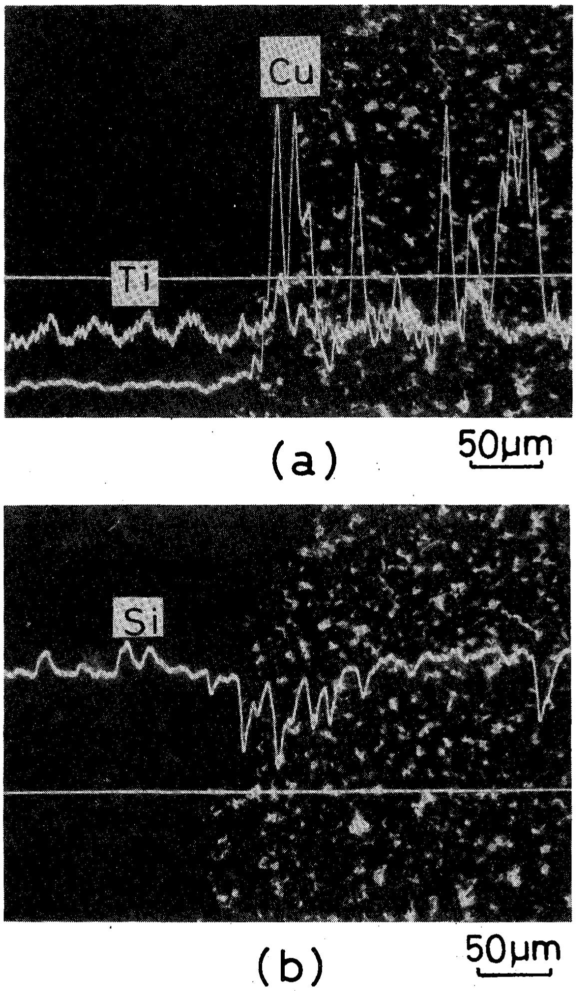

Fig. 11 Microstructures and line analyses for Ti, Cu and Si in embrittled SiC of SiC/Cu joint using $Cu_{50}Ti_{50}$ filler brazed at 1025°C for 30 min.

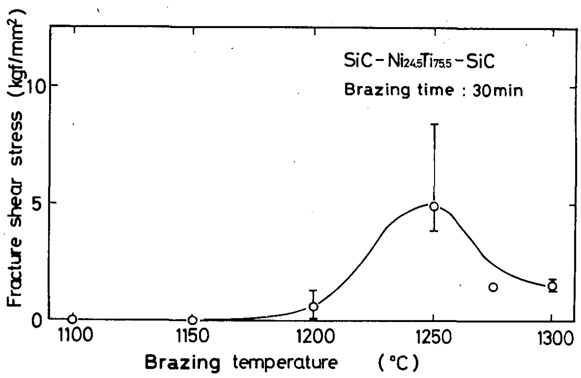

Fig. 12 Change in shear strength of SiC/SiC joint using $Ni_{24.5}Ti_{75.5}$ filler with brazing temperature at brazing time of 30 min.

3.4 Thermal stability of phases formed

Since the reaction-sintered SiC used contains the large amounts of silicon as sintering aid, the thermal stability of titanium silicides in the excess presence of silicon is thermodynamically discussed. The thermal stability of Ti silicides against Si is given by the following reactions[4].

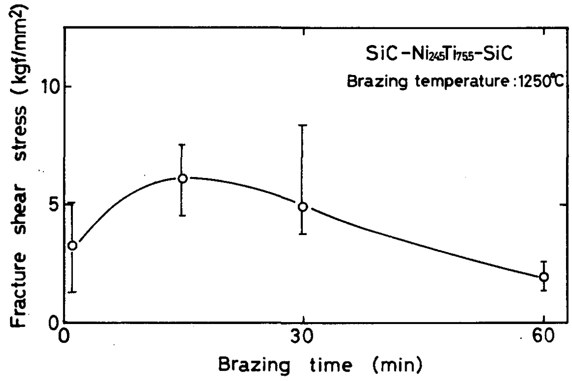

Fig. 13 Change in shear strength of SiC/SiC joint using $Ni_{24.5}Ti_{75.5}$ filler with brazing time at brazing temperature of 1250°C.

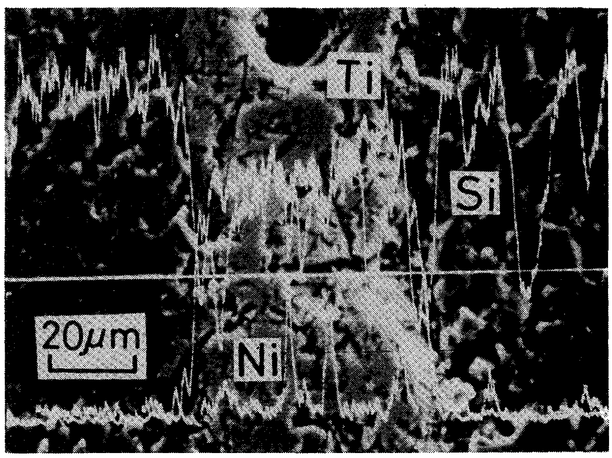

Fig. 14 Microstructure and line analyses for Ti, Cu and Si in SiC/SiC joint using $Ni_{24.5}Ti_{75.5}$ filler brazed at 1250°C for 30 min.

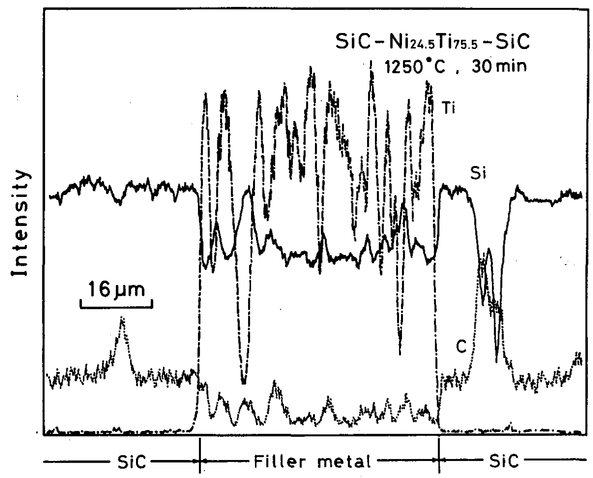

Fig. 15 Electron microprobe analyses for Ti, Si and C in SiC/SiC joint using $Ni_{24.5}Ti_{75.5}$ filler brazed at 1250°C for 30 min.

$$Ti_5Si_3(s) + 2Si(s) = 5TiSi(s) \quad (1)$$

$$\Delta G^\circ = -35175 + 11.0T$$

$$= -20072 \text{ cal/mol (at 1100°C)}$$

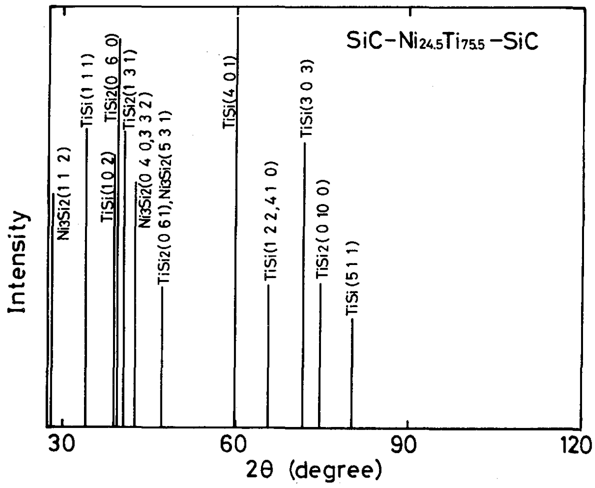

Fig. 16 X-ray diffraction pattern of fracture surface of SiC/SiC joint using $Ni_{24.5}Ti_{75.5}$ filler brazed at 1025°C for 30 min.

$$Ti_5Si_3(s) + 7Si(s) = 5TiSi_2(s) \quad (2)$$

$$\Delta G^\circ = -47805 - 9.58T$$

$$= -60958 \text{ cal/mol (at } 1100^\circ C)$$

$$TiSi(s) + Si(s) = TiSi_2(s) \quad (3)$$

$$\Delta G^\circ = -2526 - 4.12T$$

$$= -8183 \text{ cal/mol (at } 1100^\circ C)$$

Since the change of ΔG° of reactions of (1) – (3) are all negative at 1100°C, the reactions may take place during joining and the stability of Ti silicides decreases in the sequence of $TiSi_2$, TiSi and Ti_5Si_3. This supports that the Ti silicides in the brazed layer in SiC/SiC joint are composed of $TiSi_2$ and TiSi.

4. Conclusion

Joining of SiC to SiC or SiC to Cu was made using amorphous $Cu_{50}Ti_{50}$, $Cu_{43}Ti_{57}$, and $Ni_{24.5}Ti_{75.5}$ filler metals, where reaction-sintered SiC containing 13 at% Si was used. The joining strength of the lap joint of SiC/SiC or SiC/Cu joint was measured by shear fracture loading, and the joining mechanism was clarified by observing the microstructures, or analysing the elements in the joining layer. The results obtained are summarized as follows.

1) At constant brazing time of 1 min the joining strength of SiC/SiC joint exhibits a maximum value of 3 kgf/mm^2 at 1025°C using $Cu_{50}Ti_{50}$ filler, and a maximum value of 8 kgf/mm^2 at 1000°C using $Cu_{43}Ti_{57}$ filler. The joining strength of SiC/SiC joint using $Cu_{43}Ti_{57}$ filler is higher than that using $Cu_{50}Ti_{50}$ filler.
2) The joining layer of SiC/SiC joint using Cu-Ti filler is composed of $TiSi_2$, and small amount of TiSi. The main part of copper in Cu-Ti filler moves into SiC, and embrittles SiC near the joining layer of SiC/SiC joint.
3) The joining strength of SiC/Cu joint using $Cu_{50}Ti_{50}$ filler decreases from 6.5 kgf/mm^2 at brazing temperature of 1000°C to 5 kgf/mm^2 at brazing temperature of 1050°C for brazing time of 30 min. Large amounts of Cu and Ti are observed in SiC near the joining layer, and further Cu enters into SiC at a large distance from the joining layer. The copper mainly reacts with the free silicon in the reaction-sintered SiC. Thus, reaction-sintered SiC is embrittled by entering of copper. The copper melts during brazing at temperatures above 1025°C because the silicon content in copper on metal side reaches about 16 at% Si.
4) The joining strength of SiC/SiC joint using $Ni_{24.5}Ti_{75.5}$ filler exhibits a maximum value of 6 kgf/mm^2 at the brazing condition of 1025°C for 30 min, and the maximum joining strength is lower than that of SiC/SiC joint using Cu-Ti filler. At constant brazing time of 30 min the temperature for brazing SiC using Ni-Ti filler is above 1150°C, and is higher than the temperature for brazing SiC using Cu-Ti filler. The joining layer is composed of $TiSi_2$ and TiSi. The main part of Ni in Ni-Ti filler migrates into SiC during brazing of reaction-sintered SiC.

References

1) K. Yoneya: Kogyo Zairyo, 31 (1983), No. 12, 45.
2) M. Naka, K. Sampath, I. Okamoto and Y. Arata: Proc. Int. Conf. Joining of Metals, ed. by O. AL-Erhayhen, Denmark, 1984, p 169.
3) M. Naka, T. Tanaka and I. Okamoto: J. High Temp. Soc., 11 (1985), 218.
4) V. Samsonov and E. Vinitki: Handbook of High Melting Point Materials, Ni-So Tsushin Shiya, 1972, p 74.