Heat-Resistant Brazing of Ceramics (Report II)†

— Brazing of SiC to Metal Using Ni-Ti Filler Metal —

M. NAKA*, H. TANIGUCHI** and I. OKAMOTO***

Abstract

Silicon carbide SiC was brazed to Nb or Nb/W composite using Ni-50 at% Ti filler metal in vacuum. The strength and microstructure of SiC/metal joint were investigated by fracture shear loading and electron probe microanalysis, respectively. The replacement of a part of Nb with W improved the strength of SiC/Nb joint. SiC/Nb/W joint exhibited the maximum strength at a brazing condition of 1723 K and 0.6 ks, and decreased the strength at the higher brazing tempertures and longer brazing times. The excess reaction of the Ni-50 at% Ti filler with Nb prevented to react the filler with SiC, and lowered the strength of SiC/Nb/W joint. The SiC/Nb/W joint represented the high heat-resitant strength of 100 MPa at testing temperture of 1000 K. The titanium carbide TiC containing large amounts of Nb was formed at the interface between SiC and the filler

KEY WORDS: (Ceramic-Metal Joining) (Ceramic) (Silicon Carbide) (Brazing) (Filler Metals) (Niobium) (Nickel-Titanium Alloys)

1. Introduction

The joining technique of ceramics to metals provides the wide application of ceramics in various industrial fields such as automobile engine. Although the brazing technique of ceramics using Ag-Cu-Ti[1,2] and Cu-Ti[3,4] filler metals has been, in general used from the easiness of joining process, the working temperature of filler metals is limitted up to 773 K. In order to improve the heat resistance of filler metals, the Ni-Ti filler metals with higher melting points have beed used for SiC joining[5].

In the present work the silicon carbide was brazed to niobium/tumgsten composite using Ni-Ti filler metal, and the joining mechanism of joint was clarified by measuring the strength of joint and observing the microstructure of joining layer.

2. Experimental

The pressureless sintered SiC containg a few percent of Al_2O_3 was used. The metals were Nb and also W in a high purity. **Table 1** shows the physical properties of materials used. The Ni-50 at% Ti alloy was used for a filler metal. SiC in diameter of 15 mm and thickness of 3 mm of **Fig. 1 (a)** was lapped with Nb in diameter of 15 mm and thickness of 5 mm using Ni-50 at% Ti filler metal in vacuum. In order to reduce the thermal stress in SiC/metal joint the SiC/Nb joint in **Fig. 1 (b)** was brazed to W 15 mm dia. and 3 mm thick with Ni-17 at% B-8 at% filler metal at 1423 K for 0.3 ks in vacuum.

Table 1 Physical properties of SiC, Nb and W.

Materials	E (MPa)	α ($\times 10^{-6}$/K)
SiC	392000	4.2 (273-373K)
Nb	105000	7.2 (273-373K)
W	403000	4.5 (273-373K)

The brazing of SiC to Nb was performed under a load of 10 g with heating and cooling rates of 0.44 K/s in a vacuum of 1.33 mPa, The strength of joint was evaluated by fracture shear loading with a cross head spead of 1.7×10^{-2} mm/s. The reaction phases in joining layer were analysed by means of EPM analyser.

3. Results and Discussion

3.1 Joining strength of SiC/Nb/W joints

Figure 2 represents the comparison of strength for SiC/Nb joint and SiC/Nb/W joint, where SiC was brazed to Nb at 1723 K for 0.6 ks. The strength of joints was evaluated by measuring the SiC/Nb interface strength.

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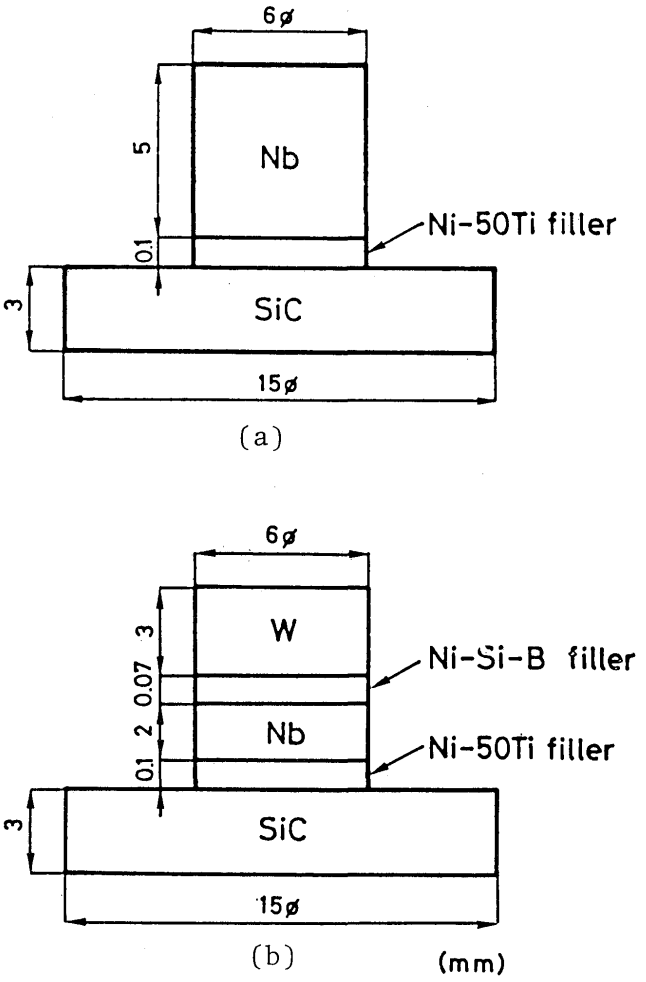

Fig. 1 Specimen size.

The replacement of a part of Nb by W effectively improves the strength of SiC/Nb joint. Although the difference in thermal expansion coefficient between SiC ($\alpha=4.2\times10^{-6}$/K) and Nb ($\alpha=7.2\times10^{-6}$/K) causes the thermal stress in SiC/Nb joint, W with the similar value ($\alpha=4.5\times10^{-6}$/K) to that of SiC reduces the thermal stress in the joint.

Figure 3 shows the brazing temperature dependence of strength for SiC/Nb/W joint at the brazing time of 0.6 ks, where the brazing is done at SiC and Nb interface. The strength of SiC/Nb/W joint exhibits the maximum value of 111 MPa at 1723 K, and decreases with increasing the brazing temperature.

The fracture surface of SiC/Nb/W joint brazed at 1723 K for 0.6 ks is shown in **Fig. 4**. The fracture structure in SiC is observed at fracture surface of SiC side and the fracture structure mixed with the interface between SiC and filler metal, and a part of SiC are observed at Nb side. The crack developes to the interface of SiC and filler metal and SiC istelf. The fracture surface of SiC/Nb/W joint brazed at 1873 K for 0.6 ks in **Fig. 5** reveales the large amounts of in-

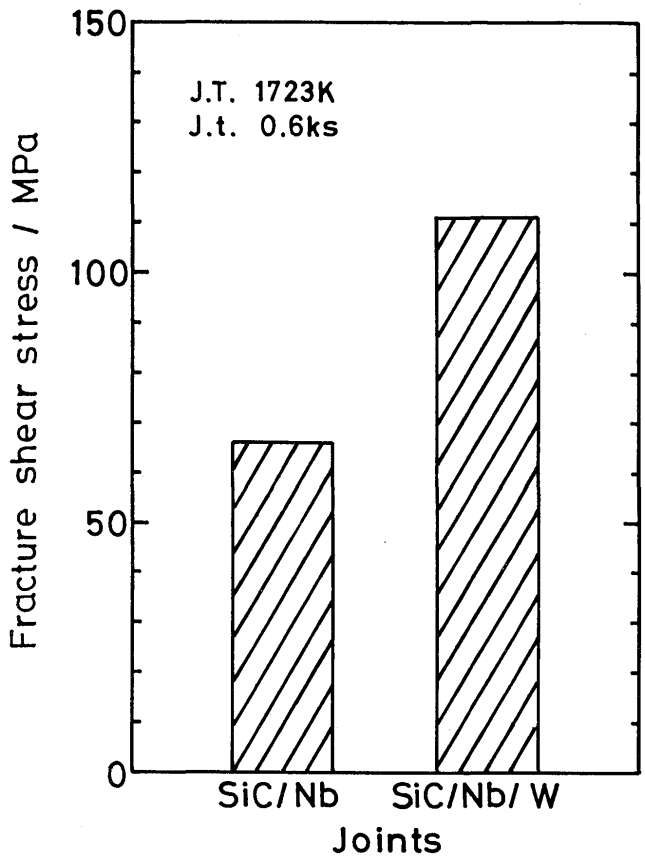

Fig. 2 Comparison of strength for SiC/Nb and SiC/Nb/W joints brazed at 1723 K for 0.6 ks.

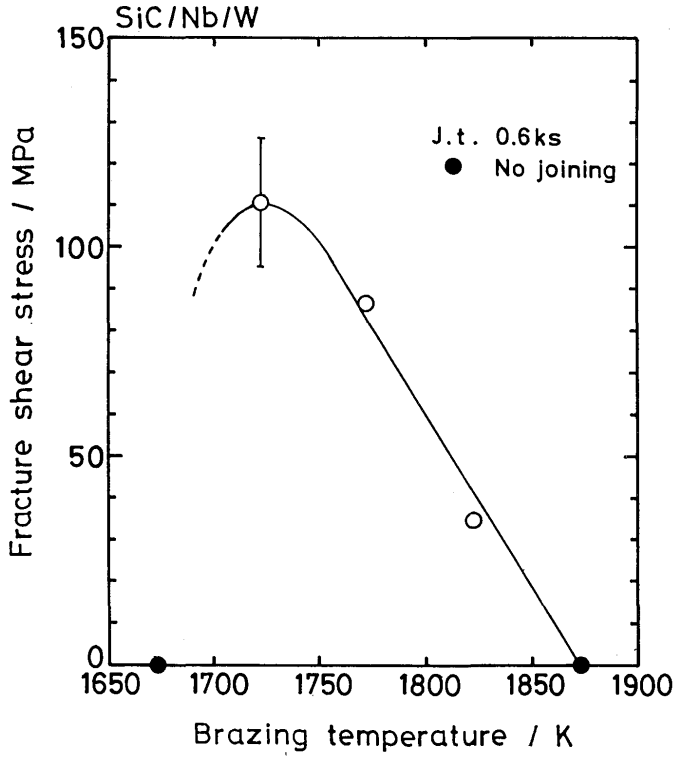

Fig. 3 Brazing temperature dependence of strength for SiC/Nb/W joint brazed at 0.6 ks.

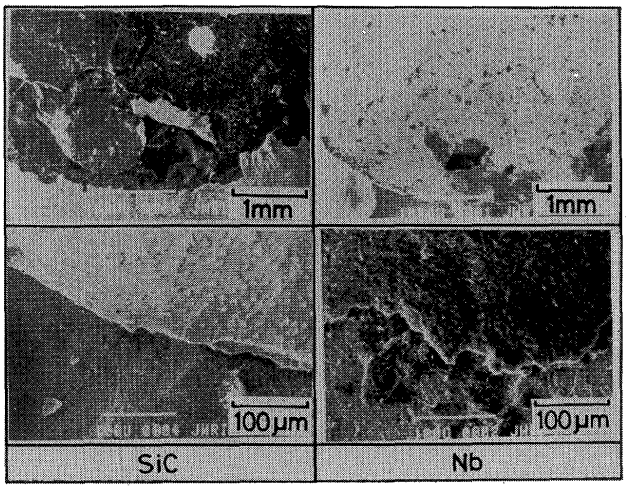

Fig. 4 Fracture surface of SiC/Nb/W joint brazed at 1723 K for 0.6 ks.

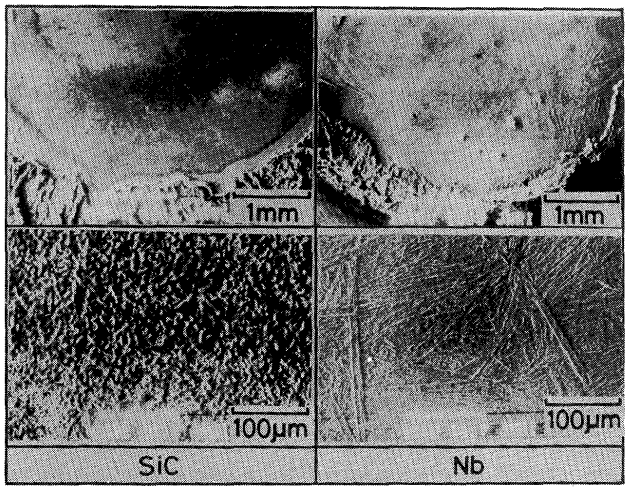

Fig. 5 Fracture surface of SiC/Nb/W joint brazed at 1873 K for 0.6 ks.

termetallic compounds in the surface at both SiC and Nb sides by the excess reaction of Ni-Ti alloy with W during brazing.

The fracture place of SiC/Nb/W joint changes from the mixture of SiC ceramic and the filler metal near the interface at 1723 K to only the filler metal at 1873 K. This transition of fracture place in the joint implies that the excess reaction of Ni-50 Ti alloy with W at 1873 K prevents the reacting and joining of Ni-Ti alloy with SiC.

The effect of brazing time on the strength of SiC/Nb/W joint brazed at 1723 K is shown in **Fig. 6**. The strength of joint lowers monotonously with increasing brazing time. SiC was not joined to Nb at joining time of 3.6 ks. The fracture places of joint changes from the mixture of interface, SiC and filler metal to interface and filler metal. The excess formation of intermetallic compounds between the filler metal and W reduces the

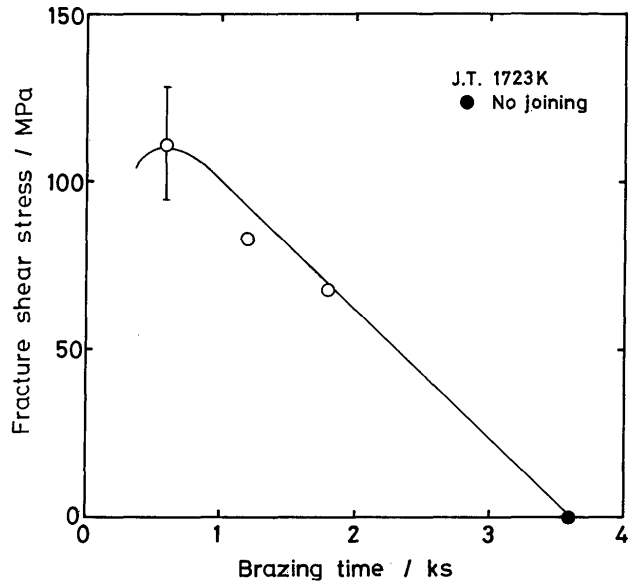

Fig. 6 Brazing time dependence of strength for SiC/Nb/W joint brazed at 1723 K.

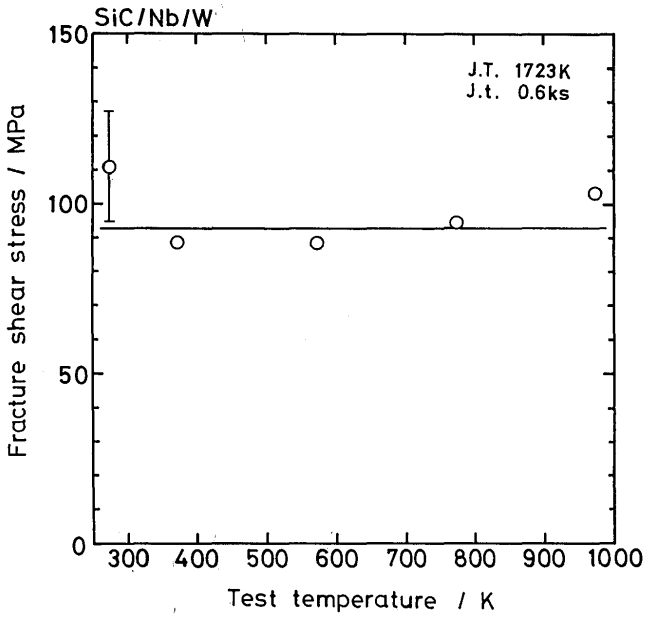

Fig. 7 Change in strength of SiC/Nb/W joint with testing temperature.

strength of SiC/Nb joint.

Figure 7 shows the change in strength of SiC/Nb/W joint with testing temperature, where SiC was brazed at 1723 K for 0.6 ks. The joint represents no temperature dependence of strength at testing temperature up to 1000 K, and shows the high strength of 100 MPs at 973 K. The fracture places of joints are SiC itself or interface of joint at all testing temperatures from room temperture to 973 K as shown in **Fig. 8**. The observation of fracture surface for the SiC joints indicates the crack developes into SiC itself since the strength of SiC/Nb interface is high.

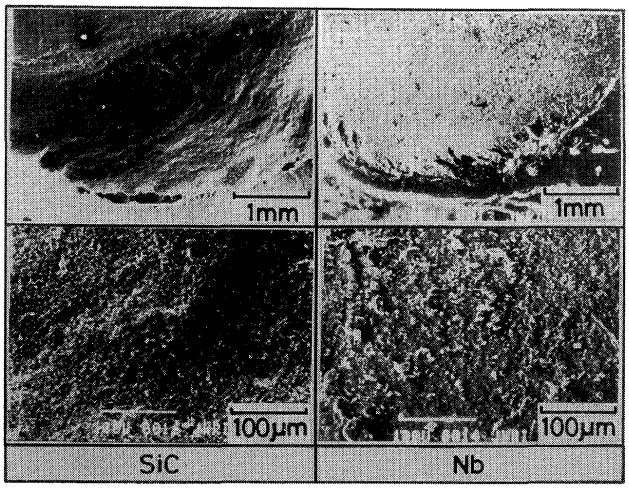

Fig. 8 Fracture surface of SiC/Nb/W joint at testing temperature at 973 K.

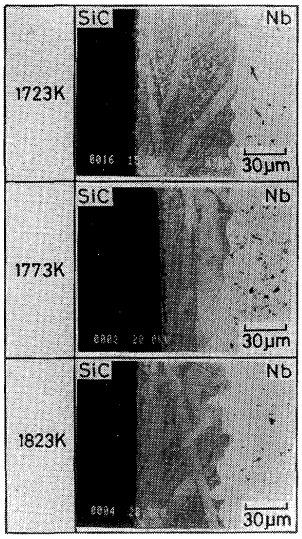

Fig. 9 Change in microstructure of SiC/Nb/W joint brazed for a 0.6 ks with brazing temperature.

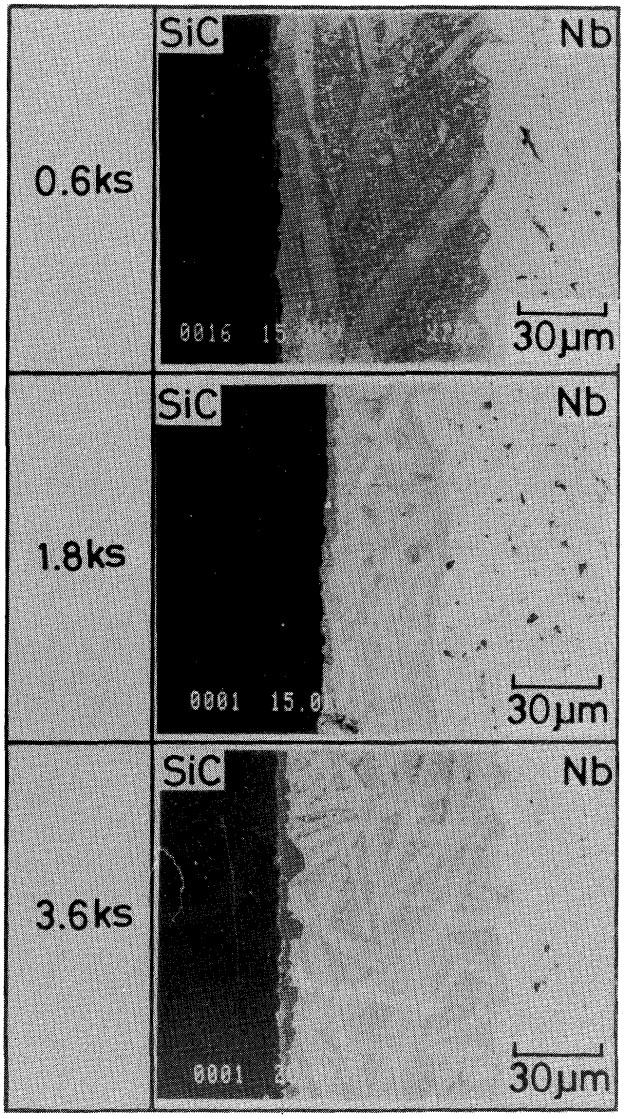

Fig. 10 Change in microstructure of SiC/Nb/W joint brazed at 1723 K with brazing time.

3.2 Microstructure of SiC/Nb/W joint

Figure 9 shows the change in microstructure of SiC/Nb interface in SiC/Nb/W joint brazed at 0.6 ks with brazing temperture. The thickness of thin layer carbide (Ti, Nb)C which is identified by EPM analyer described later increases a little with increasing the brazing temperture. The primary lief-like phases which is identified as Nb-Ni-Ti ternary solid solution crystallize in the filler. The increase in the size of the crystallized phases with increasing the brazing temperature is attributable to the degradation of SiC/Nb/W joint.

The increase in the amount of primary phases in the filler with increasing the brazing time at a brazing temperture of 1723 K also degrades the strength of SiC/Nn/W joint as shiwn in **Fig. 10**. The microstructure and X-ray image analysis of Ti, Ni, Nb, Si and C are shown in **Fig. 11**. The X-ray image analyses indicate that the thin layer phase at the interface between SiC and the filler contains Ti, Nb and C elements, and is identified as (Ti, Nb)C by the quantitative analyses described later.

Figure 12 and **Table 2** show the microstructure and quantitative analyses of phases in SiC/Nb interface of joint brazed at 1723 K for 0.6 ks, respectively. The layer phase at SiC/filler metal interface is identified as (Ti, Nb)C Carbide, where the amount of 30 atomic percent for titanium is replaced by niobium. During brazing nibium dissolves into the filler, and the (Ti-Nb-Vi-Si) (iii) and (Ti, Nb)Ni (iv) phases are solidified as the primary phases from the filler. Furthermore, $(Ti, Nb)_2Ni$ (ii) besides SiC, and (Ni-Ti-Nb) (v) and (Nb-Ti-Ni) (iv) phases in (Ni-Ti-Nb) eutertic structure (vii) are observed in the filler. TiC and NbC are completely dissolved because the two carbides possess the same lattice structure of cubic, Bl type.

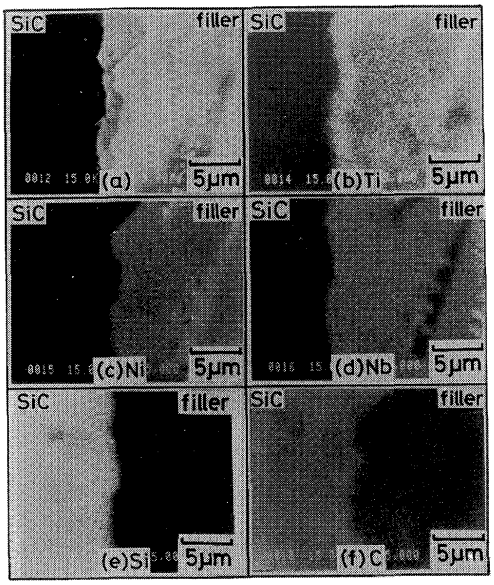

Fig. 11 Microstructure and X-ray image analyses of Ti, Ni, Nb, Si and C in SiC/Nb/W joint brazed at 1723 K for 0,6 ks.

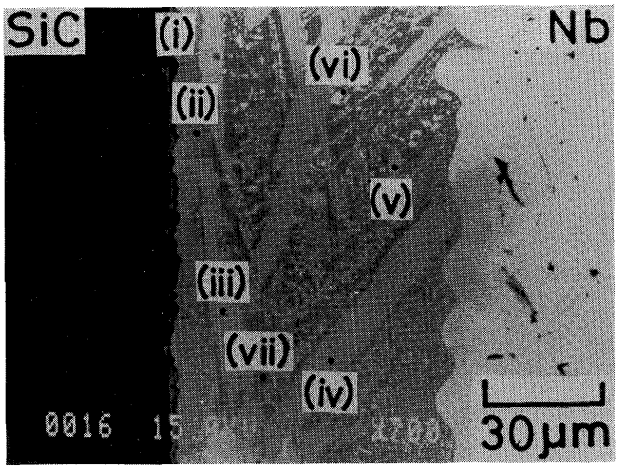

Fig. 12 Microstructure of SiC/Ni-50Ti/Nb interface brazed at 1723 K for 0.6 ks.

Table 2 Quantitative analyses of joining layer at SiC/Nb interface for SiC/Nb/W joint brazed at 1723 K for 0.6 ks.

Phase	Composition (at%)					
	C	Ti	Nb	Ni	Si	Al
(i)	46.5	41.1	10.7	1.4	0.2	0.0
(ii)	—	41.8	24.6	32.5	1.2	0.1
(iii)	—	17.3	22.9	17.3	16.6	0.0
(iv)	—	28.7	33.8	33.6	3.4	0.6
(v)	—	32.5	24.9	37.6	3.3	1.7
(vi)	—	23.8	64.1	11.7	0.1	0.5
(vii)	—	46.8	5.4	47.5	0.1	0.5

The change in Nb/Ti ratio of (Ti, Nb)C carbide at the interface between SiC and filler is shown in **Fig. 13**. The niobium content in the carbide extremely rises from Nb/Ti=0.3 at 1723 to Nb/Ti=0.75 at 1823 K.

The microstructure and X-ray image analyses of Nb/W interface in SiC/Nb/W joint brazed at 1423 K for

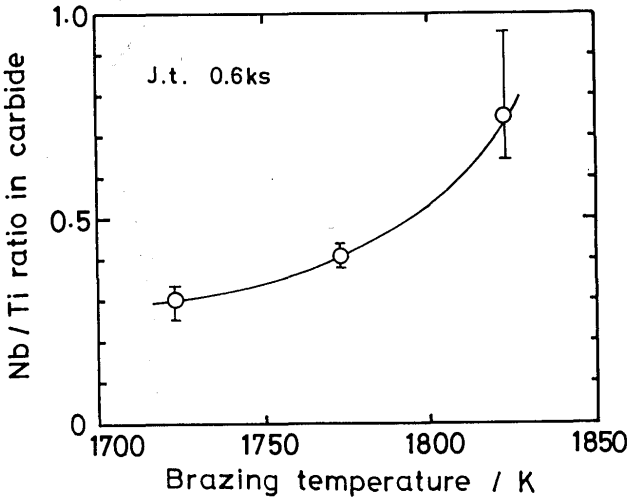

Fig. 13 Change in Nb/Ti ratio of carbide for SiC/Nb/W joint brazed for 0.6 ks.

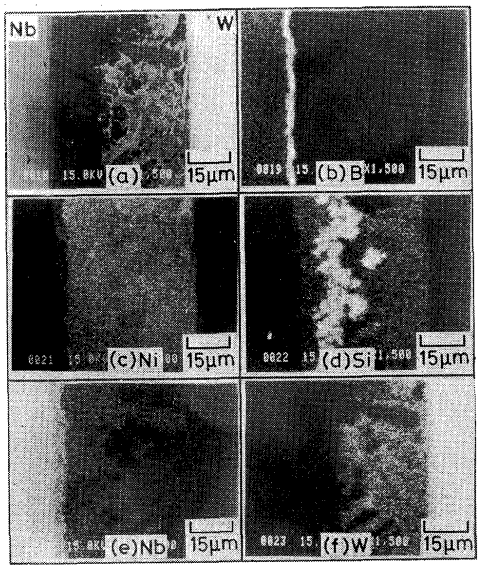

Fig. 14 Microstructure and X-ray image analyses of B, Ni, Si, Nb and W at Nb/W interface in SiC/Nb/W joint brazed at 1423 K for 0.3 ks.

0.3 ks is shown in **Fig. 14**. The Nb and B rich layer phase containing Ni is observed at SiC/filler metal interface. The ratio of Ni/Nb is about 0.5 as shown in Fig. 15 and Table 3, where the quantitative analyses of element is done by EPMA analyser. The layer phase contains 8.7 at% B. At the filler metal/W interface $(Ni, W)_3B_2$ phase containing large content of 26.9 at% W is observed. In the filler metal Ni_2B_3 (ii), Ni_3(Nb, Si) (iii) phases beside Nb and (Nb-Ni-Bi-Si) (iv) and (Ni-W-Nb-B-Si) phases beside W are observed as shown in **Fig. 15** and **Table 3.**

3.3 Reaction mechanism of Ni-Ti alloy with SiC

The growth rate of carbide formation at the interface between SiC and Ni-Ti alloy is approximated by the Fick's law as,

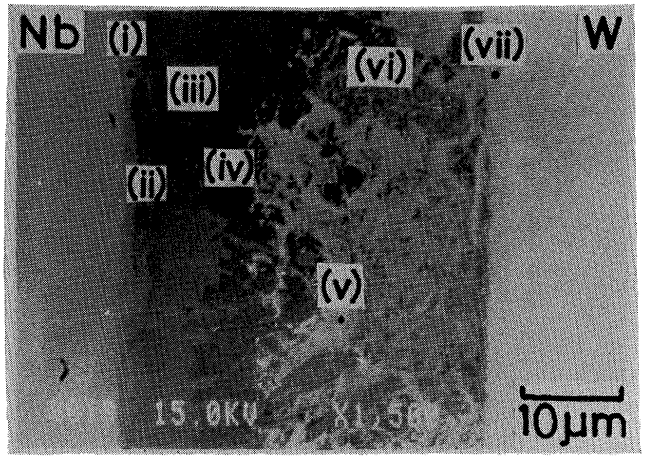

Fig. 15 Microstructure of Nb/Ni-Si-B/W interface brazed at 1423 K for 0.3 ks.

Table 3 Quantitative analyses of joining layer at Nb/W interface for SiC/Nb/W joint at 1423 K for 0.3 ks.

Phase	Composition (at%)				
	Nb	Ni	B	Si	W
(i)	49.7	40.7	8.7	0.9	0.0
(ii)	24.7	18.4	56.3	0.6	0.0
(iii)	19.7	74.6	0.0	5.0	0.7
(iv)	14.2	68.5	6.3	10.6	0.4
(v)	9.9	46.0	23.9	2.8	17.4
(vi)	23.9	60.4	5.2	6.6	3.4
(vii)	6.9	25.2	41.0	0.0	26.9

$$X^2 = kt \tag{1}$$

$$k = k_0 t \exp(-Q/RT) \tag{2}$$

$$X^2 = k_0 t \exp(-Q/RT) \tag{3}$$

$$2\,lnX = lnk_0 + lnt - Q/RT \tag{4}$$

For the brazing time is fixed as 0.6 ks, the activation energy of Q is obtained from the relation of 2 lnX against 1/T in **Fig. 16**, where the date for SiC/Ni-50Ti/SiC and for SiC/Ni-50Ti/Nb are plotted by the symbol marks of ○ and △, respectively. The activation energies for both systems are 261 KJ/mol, which is similar value to the value of 271 kJ/mol for Ti/SiC solid system[6]. The formation of TiC in Ni-Ti/SiC system is dominated by the same reaction of Ti and SiC in Ti/SiC system. The growth of TiC carbide in SiC/Nb system is higher than that in SiC/SiC system. The dissolution of Nb to TiC affects the factor of k in eq. (3) and retards the formation of TiC.

4 Conclusion

The brazing of SiC to Nb or Nb/W composite was performed using Ni-50 at% Ti filler metal in vacuum. The joining mechanism was investigated by measuring the fracture shear loading, and observing the microstructure of joint using EPM microanalyser.

The replacement of a part of Nb with W definitely

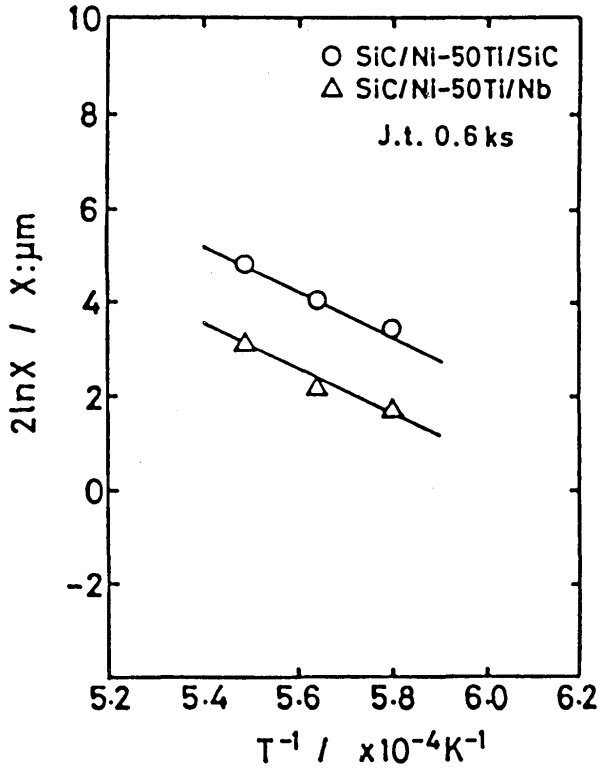

Fig. 16 2ln X plotted with 1/T at a brazing time of 0.6 ks in SiC/SiC and SiC/Nb/W joints with Ni-50Ti filler, where X and T are thickness of carbide and brazing temperture, respectively.

improves the strength of SiC/Nb joint, where W is brazed with Ni-Si-B filler metal.

The brazing condition of 1723 K and 0.6 ks gives the maximum strength of SiC/Nb/W joint, and the higher brazing temperature and longer brazing time provides the lower strength of joint. The excess reaction of Ni-50 at% Ti alloy with Nb prevents the reaction of Ni-50 at% Ti alloy with SiC and degrades the strength of SiC/Nb/W joint. The SiC/Nb/W joint represents the high heat-resistant strength of 100 MPa at testing temperature of 1000 K. The formation of TiC at SiC/Ni-50 at% Ti filler is attributable to the high strength of SiC/Nb/W joint. The dissolution of Nb to TiC retards the growth of TiC carbide, though the activation energy for growth of TiC is the same value of 271 KJ/mol as that of SiC/Mi-50Ti interface.

References

1) H. Takeda: Trend in Joining Technology, Japan Weld Soc., (1986), 32.
2) T. Yano, H. Suematsu and T. Iseki: J. Mater. Sci., 23 (1988), 3362.
3) M. Naka, T. Tanaka and I. Okamoto: J. High Temp. Soc., 11 (1985), 218.
4) M. Naka and I. Okamoto: Materials Science and Engineering, 98 (1988), 81.
5) M. Naka, H. Taniguchi and I. Okamoto: Trans. of JWRI, 19 (1990), No. 2, 25.
6) K. Kurokawa and R. Nagasaki: Sintering 87, Elsevier Applied Science (1987), 1397.