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Bonding Behavior between Niobium and Reaction-Sintered SiC†

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Abstract

Reaction-sintered SiC containing 13 mass% Si is joined to Nb by solid state bonding in a vacuum. The joining strength of SiC/Nb joint increases to the saturated value of 88 MPa with increasing the joining temperature at the joining time of 1.8 ks and the joining pressure of 1.96 MPa. An increase in joining pressure raises the joining area at the interface of SiC/Nb joint. The change in joining time changes a little the joining strength of the joint at the constant joining temperature and pressure since the fracture takes place at the interface between the intermediate phase of Nb_5Si_3 and Nb. The intermediate phase is Nb_5Si_3 at joining temperatures below 1677 K. The growth kinetic is expressed by Fick's law, and the activation energy for growth of Nb_5Si_3 is 456 kJ/mol. The growth of $NbSi_2$ is preferential at joining temperatures above 1773 K.

KEY WORDS : (Joining) (Solid State Bonding) (Ceramics) (Silicon Carbide) (Niobium) (Niobium Silicide)

1. Introduction

Joining of ceramics to metals is necessary for the application of ceramics. The joining methods are divided into the solid state bonding, brazing and metallizing methods. Several works on brazing of ceramics to metals have been reported; the utilizing of amorphous Ti-base filter[1,2] and aluminum alloy filler[3], and the utilizing of metal-oxide[4]. Several investigations on joining of SiC to metals have been reported using Al filler[5], Cu-Mn filler[6] and Ag-Cu-Ti filler[7].
These reports were, however, focused on the brazing methods using the filler metals.

On the other hand, the solid state bonding provides the heat-resistant strength of joint and easiness of reaction control in the joining interface, compared with other joining methods such as brazing process. The present investigation is concerned with the solid state bonding of reaction-sintered SiC with free-silicon to Nb which possesses the low thermal expansion coefficient of $7.2 \times 10^{-6}/$ K and clarifies the change in joining strength of the SiC with joining conditions such as joining temperature, joining time and joining pressure, and also the phase reactions during joining.

2. Experimental

Reaction-sintered SiC (R.S.SiC) used contains the free

Si of 13 % as sintering aids. Electron-beam melting Nb used contains 0.01 % O. SiC in 6 mm diameter and 4 mm length was joined to Nb in 6 mm diameter and 4 mm length in the joining conditions of 1373-1773 K, 1.8-21.6 ks and 0.49-1.96 MPa under a vacuum condition of 13.3 mPa. The construction of heating part was given in elsewhere[8]. The temperature and pressure hysteresis is shown in **Fig. 1**. The joining was conducted in a vacuum condition of 1.33 mPa with a heating and cooling rate of 0.93 K/s. The joining strength was evaluated by fracture shear stress with a cross head speed of 1.67×10^{-2} mm/s. The joining interface was investigated by means of scanning electron microscope, EDX microanalyser and X-ray diffractometer.

3. Results and Discussion

3.1 Joining strength of SiC/Nb joint

Figure 2 shows the joining temperature dependence of fracture strength for R.S.SiC/Nb joint joined at the joining time of 1.8 ks under 0.49 MPa or 1.96 MPa. The strength of R.S.SiC/Nb joint increases with increasing the joining temperature under the joining pressure of 0.49 MPa. The SEM micrograph of fracture surface of R.S.SiC/Nb joint is given in **Fig. 3**, where the joint was
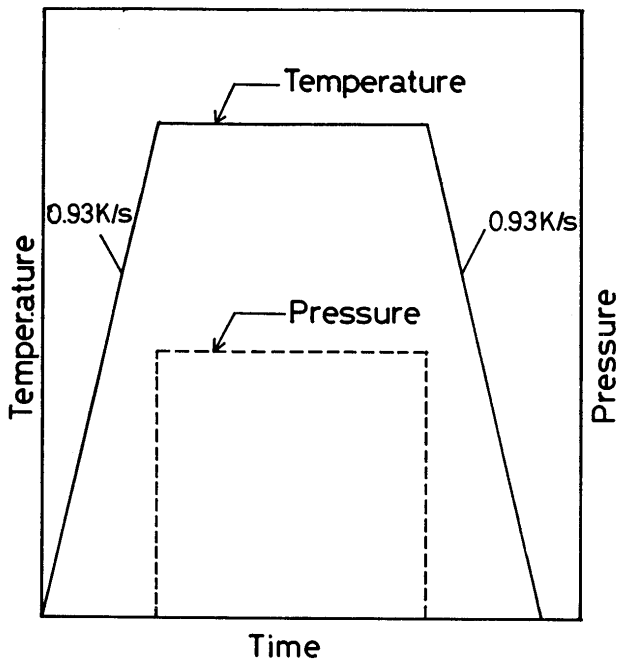
Fig. 1 Temperature and pressure hysteresis during joining.
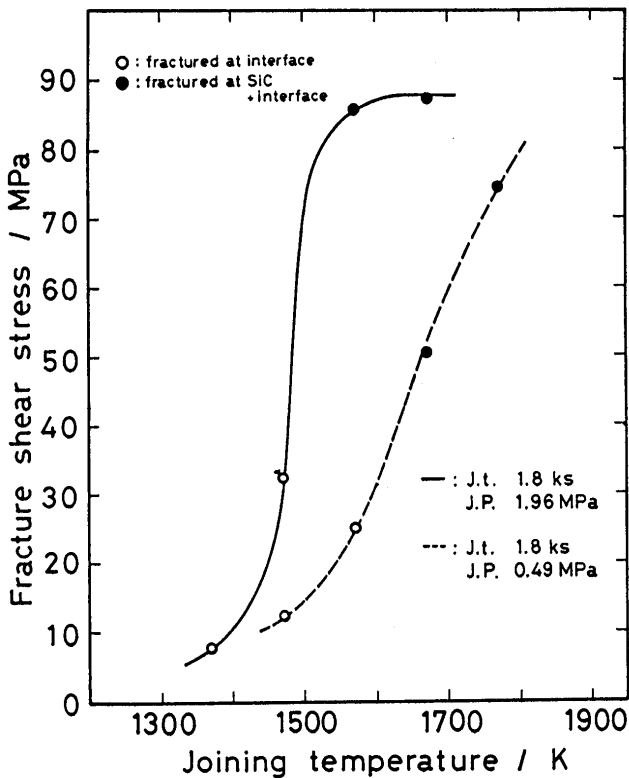

Fig. 2 Joining temperature dependence of fracture strength for R.S.SiC/Nb joint joined at 1.8 ks under 0.49 MPa (---), and 1.8 ks under 1.96 MPa (—) where the fracture takes place at interface of joint (○) or at SiC and interface (●).

joined at 1573 K for 1.8 ks under 0.49 MPa. The fracture surface of Nb part and SiC part are shown in the figure. The fracture takes place at the interface of the joint at the joining temperatures below 1573 K where the joining strength is relatively low. The fracture surface of R.S.SiC/Nb joint is shown in **Fig. 4**, where the joint was joint at 1673 K for 1.8 ks under 0.49 MPa. The fracture takes place at SiC itself plus interface at higher joining temperatures. Under the joining pressure of 1.96 MPa the strength of SiC/Nb joint rises steeply with increasing the joining temperature, and reaches the saturated value of 88 MPa.

The joining time dependence of fracture strength for R.S.SiC/Nb joint is shown in **Fig. 5**. The strength of the joint shows only a little change against the joining time because the fracture of joints takes place at the interface
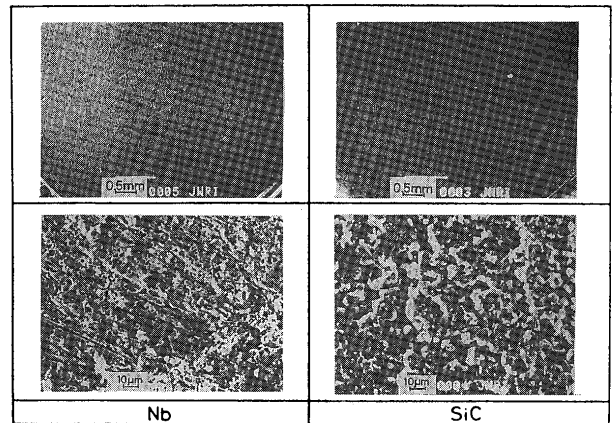

Fig. 3 Fracture surface of R.S.SiC/Nb joint at room temperature, where the joint was joined at 1573 K for 1.8 ks under 0.49 MPa.
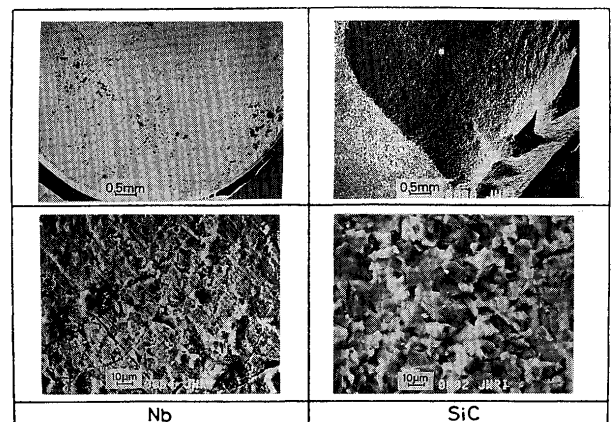

Fig. 4 Fracture surface of R.S.SiC/Nb joint at room temperature, where the joint was joined at 1673 K for 1.8 ks under 0.49 MPa.

between the intermediate phase and Nb.

In **Fig. 6** the joining strength of R.S.SiC/Nb joint effectively increases from 38 MPa to 87 MPa with increasing the joining pressure from 0.49 MPa to 1.96 MPa at the joining condition of 1673 K and 1.8 ks. The joining pressure definitely increases the joining area at the joining interface. This leads to the increase in joining strength with the joining pressure in Fig. 6.

The fracture strength of R.S.SiC/Nb joint joined at 1673 K for 1.8 ks under 1.49 MPa was investigated at the elevated temperatures from room temperature to 1073 K as shown in **Fig. 7**. The SiC/Nb joint shows only the gradual decrease of the strength against the elevated temperatures. Even at the testing temperature of 1073 K the SiC/Nb joint possesses the strength of 52 MPa. The melting point of Nb-Nb$_5$Si$_3$ system, which was identified later at the joining interface of SiC/Nb joint, is above 2153 K[9]. Further the fracture surfaces of Nb part and SiC part in
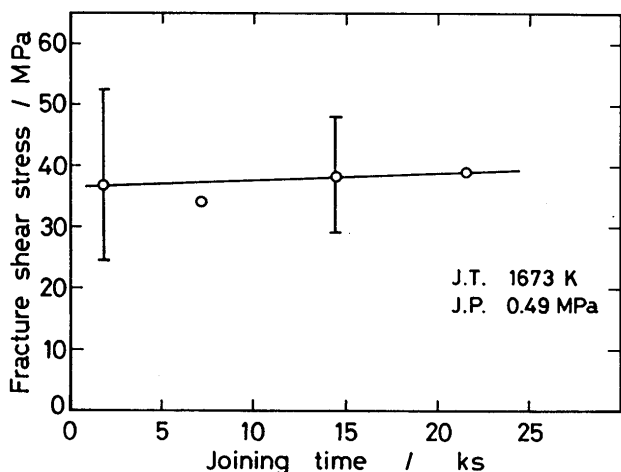

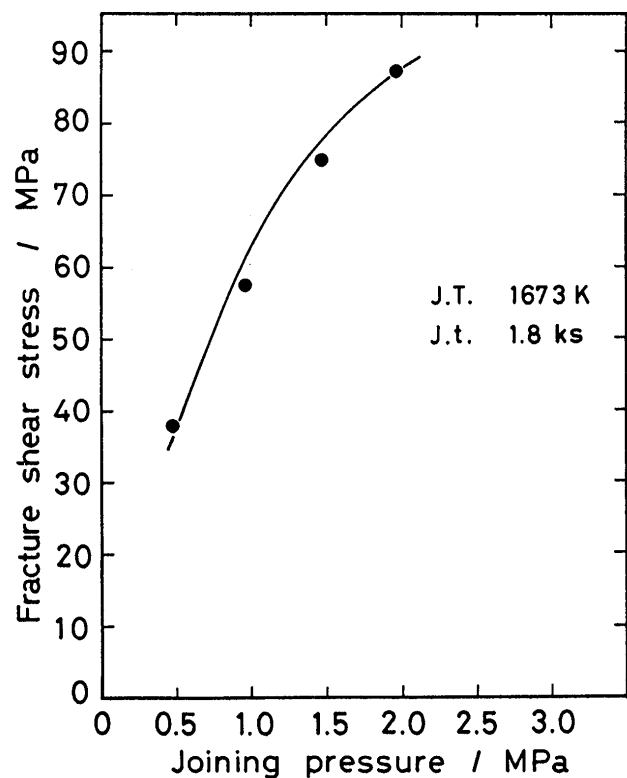Fig. 5 Joining time dependence of fracture strength for R.S.SiC/Nb joint at 1673 K under 0.49 MPa.

Fig. 6 Joining pressure dependence of fracture strength for SiC/Nb joint joined at 1673 K for 1.8 ks.
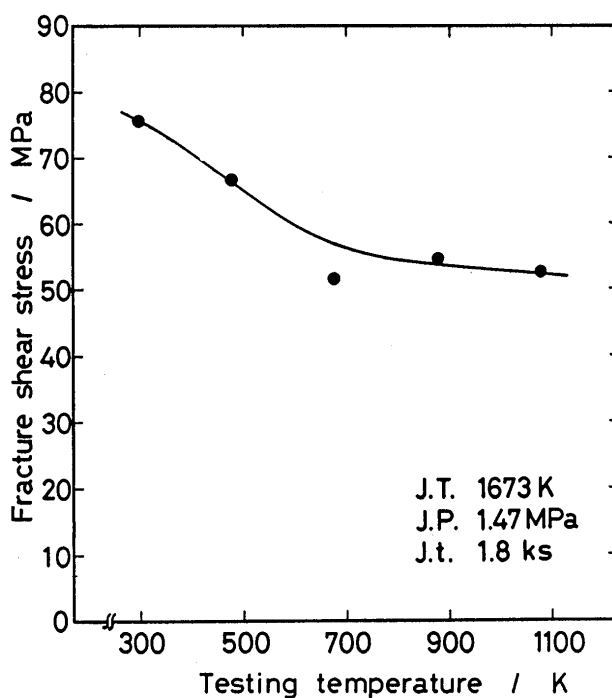

Fig. 7 Effect of elevated temperature on fracture strength for R.S.SiC/Nb joint joined at 1673 K for 1.8 ks under 1.49 MPa.
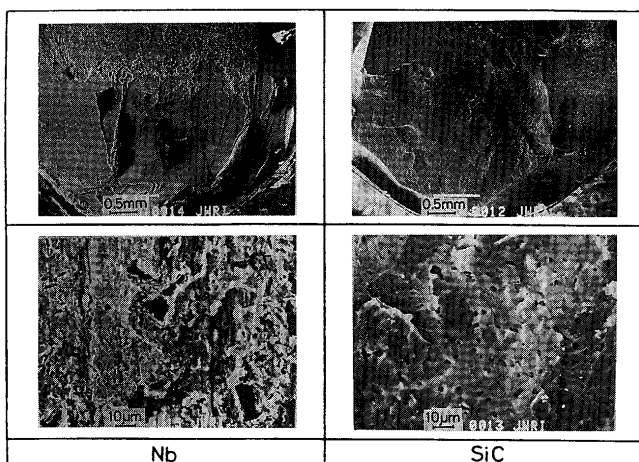

Fig. 8 Fracture surface of R.S.SiC/Nb joint at 1073 K, where the joint was joined at 1673 K for 1.8 ks under 0.49 MPa.

R.S.SiC/Nb joint at the testing temperature of 1073 K show the similar structure to that of the joint fractured at room temperature as shown in **Fig. 8**. These facts account for the high heat-resistant property of R.S.SiC joint in Fig. 7.

3.2 Intermediate phases at interface between SiC and Nb

The microstructural analyses were performed using energy dispersive X-ray (EDX) micronalysis and scanning electron microscopy. **Figure 9** shows the scanning electron microstructure, and line analyses of silicon and niobium (a) of the R.S.SiC/Nb joint at 1573 K for 21.6 ks under 0.49 MPa. The intermediate phase (i) in Fig. 9(a) is identified as niobium silicide (Nb_5Si_3) from the EDX spot analyses in Fig. 9(i) and the standard specimens of niobium silicides. **Figure 10** shows the scanning electron microstructure and line analyses of Si and Nb in the R.S.SiC/Nb joint joined at 1673 K for 14.4 ks under 0.49 MPa. The intermediate phases (i) and (ii) are identified as Nb_5Si_3 and $NbSi_2$, respectively from EDX spot analyses in Fig. 10 (i) and (ii), and the standard specimens of

niobium silicides. The intermediate phase is initially Nb_5Si_3 at a joining time of 3.6 ks, and it becomes two silicides at longer times at 1673 K. Further, at the higher joining temperature of 1773 K (**Fig. 11**), the preferential growth of $NbSi_2$ is dominant though the two silicides Nb_5Si_3 and $NbSi_2$ are formed in the intermediate phase as shown in the line and spot analyses of Nb and Si in R.S.SiC/Nb joint.
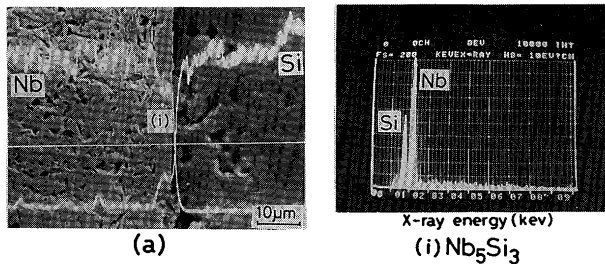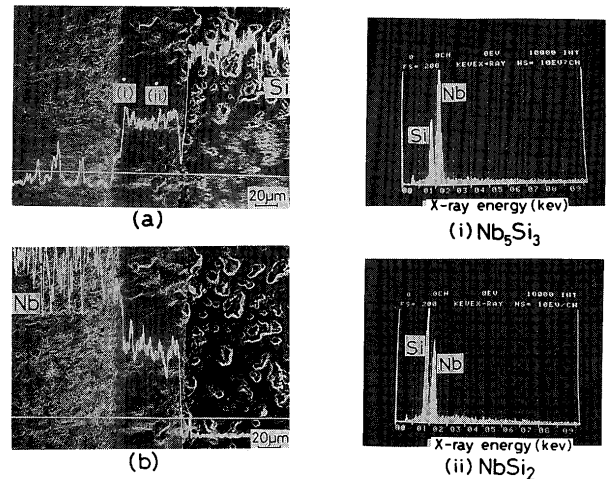

Fig. 11 Line analyses of Nb (a) and Si (b) and spot analyses (i,ii) of Nb_5Si_3 and $NbSi_2$ in R.S.SiC/Nb joint joined at 1773 K for 21.6 ks under 0.49 MPa.

Fig. 9 Line analyses of Nb and Si (a) and spot analysis (i) of Nb_5Si_3 (b) in R.S.SiC/Nb joint joined at 1573 K for 21.6 ks under 0.49 MPa.
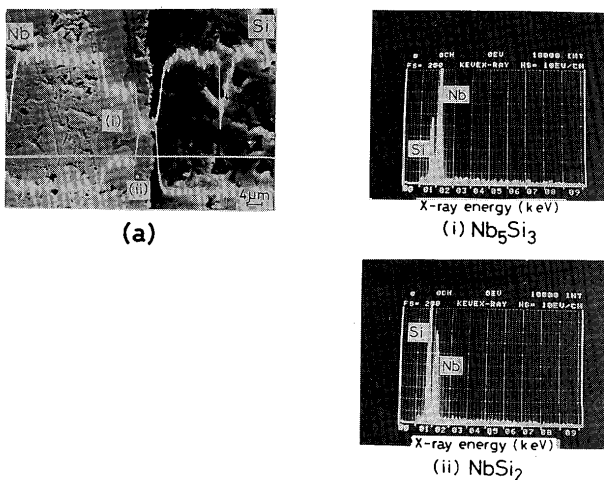

Fig. 10 Line analyses of Nb and Si (a) and spot analyses (i,ii) of Nb_5Si_3 and $NbSi_2$ in R.S.SiC/Nb joint joined at 1673 K for 21.56 ks under 0.49 MPa.
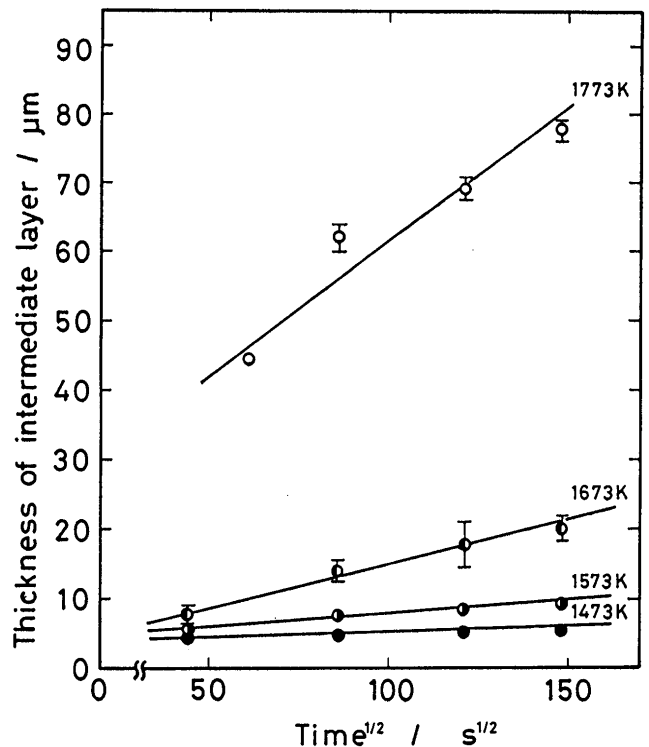

Fig. 12 Thickness of intermediate phase for SiC/Nb plotted against square root of joining time under 0.49 MPa.
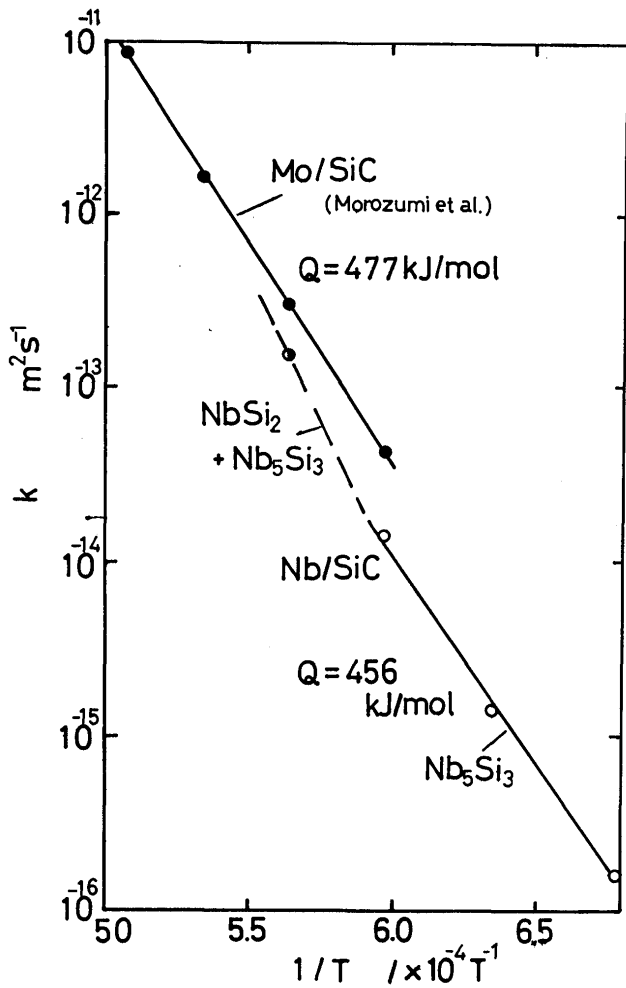
Fig. 13 k plotted against 1/T, where k is the rate constant of intermediate phases, ($\bigcirc$) for Nb_5Si_3 and ($\bullet$) for $NbSi_2 + Nb_5Si_3$.

At joining temperatures below 1673 K the intermediate phase is Nb_5Si_3, and the growth of Nb_5Si_3 is represented by Fick's law as shown in **Fig. 12**.

$$d^2 = kt \qquad (1)$$

where d, t and k are the thickness of Nb_5Si_3, joining time and rate constant, respectively. Further, the activation energy for growth of Nb_5Si_3, Q is expressed by (**Fig. 13**),

$$k = k_0 exp(-Q/RT) \qquad (2)$$

From the slope in Fig. 13, Q is estimated to be 456 kJ/mol. This value for SiC/Nb is a little lower than that of 477 kJ/mol for SiC/Mo system[10]. Large amounts of free silicon contained in the SiC for the present work may promote the formation of the silicides. At the higher joining temperatures of 1773 K, the preferential growth of $NbSi_2$, was formed as the intermediate phase at the inter-

face SiC/Nb joint. This result indicates that the thermal stability of silicides changes from Nb_5Si_3 to $NbSi_2$ with increasing the joining temperature.

4. Conclusion

The joining of reaction-sintered SiC to Nb was conducted at the joining conditions of 1373-1773 K, 1.8-21.6 ks and 0.49-1.96 MPa under a vacuum condition of 13.3 mPa.

The fracture strength of SiC/Nb joint increases with increasing with increasing temperature at the joining time of 1.8 ks under the joining pressure of 0.49 or 1.96 MPa.

The joining between R.S.SiC and Nb begins at the joining temperatures above 1373 K, and the strength of the joint reaches the saturated value of 88 MPa with increasing joining temperature at 1.96 MPa.

The increase in joining pressure raises the joining strength of SiC/Nb joint at the constant joining temperature and time. This indicates that the increase in joining area leads to the increase in the joining strength of the joint.

The intermediate phase Nb_5Si_3 at the joining temperatures below 1677 K is grown by Fick's law, and the activation energy for the growth is 456 kJ/mol. At the joining temperature of 1773 K the growth of $NbSi_2$ is preferential.

References

1) M. Naka, K. Sampath and I. Okamoto: Engineering Application & Ceramic Materials, ASM, 1985, p205.

2) M. Naka, T. Tanaka and I. Okamoto : Quaterly J. Japan Weld. Soc., 5(1986), 113.

3) M. Naka, Y. Hirono and I. Okamoto : Quaterly J. Japan Weld. Soc., 5(1987), 31.

4) M. Naka and I. Okamoto: Quaterly J. Japan Weld. Soc., 3(1985), 702.

5) A. Kohno, T. Yamada and K. Yoko, J. Japan Inst. Metals, 49(1985), 876.

6) H. Okamura, K. Miyazaki, T. Matzuzaki, T. Shida, M. Ura and T. Okuo : Quaterly J. Japan Weld. Soc., 4(1986), 477.

7) T. Izeki, H. Matsuzaki and J. K. Boadi : Am. Ceram. Soc. Bull., 64(1985), 322.

8) M. Naka and I. Okamoto : Trans. JWRI, 15(1986), No. 1, 157.

9) M. Hansen: "Constitution of Binary Alloys" 2nd ed. 1985, p1016.

10) S. Morozumi, M. Kikuchi, S. Sugai and M. Hayashi : J. Japan Inst. Metals, 44(1980), 1404.