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Interfacial Structure and Reaction Mechanism of SiC/Nb Joints†

Jicai. FENG*, Masaaki. NAKA** and Julius. C. SCHUSTER***

Abstract

SiC was joined to SiC using Nb foil at temperatures ranging from 1116 to 1790 K for 1.8~36 ks and 7.26 MPa in vacuum. Interfacial reactions and microstructures were investigated using electron probe microanalyses and X-ray difractometer. SiC begins to join with Nb at temperatures above 1116 K. At 1678 K for 14.4 ks, hexagonal Nb_2C phase is formed at the Nb side of the reaction zone, while the hexagonal Nb_5Si_3C phase is formed at the interface between Nb_2C and SiC. The same reaction structure can be observed at 1790 K for 1.8 ks. Cubic NbC phase was formed at two interfaces of SiC/Nb_5Si_3C and Nb_2C/NbC at 1790 K for 36 ks.

The thickness of two reaction layers are increased with the bonding temperature and also with bonding time. The reaction layers grow by Fick's law and the activation energies for growth are 382 and 353 kJ/mol for Nb_5Si_3C and Nb_2C, respectively.

KEY WORDS: (Ceramics/Metal Interface) (Solid State Bonding) (Reaction Mechanism) (Interface Structure) (Activation Energy) (Niobium Carbide) (Reaction Layer) (Growth Kinetics)

1. Introduction

Silicon carbide is one of the candidates for the high temperature structural components used up to 1273 K with its remarkable features such as light weight, high strength and good resistance to wear. Some structural application of ceramics requires a reliable joining method with a metallic component. Compared to other joining methods such as brazing, solid state bonding provides high temperature strength of the joint as well as relatively easy control of the interfacial reaction. However, for effective control of the interface structure, study of the phase reactions between the ceramic and the metal is necessary.

While the interfacial structures, phase reactions, and the formation of titanium carbide and silicides at the SiC/Ti interface were investigated in detail[1-4], few reports exist on the SiC/Nb system. According to Naka[5,6], Nb_5Si_3 and Nb_2C were formed in the reaction zone at temperatures from 1473 K to 1773 K. On the other hand, Joshi et al.[7] observed phase reactions in thin Nb film on SiC, but did not characterize microstructures of observed phases in the system.

The aim of the present work is to examine the interfacial structure, phase reaction, growth of layers, and diffusion path between SiC and Nb.

2. Materials and experimental procedure

The materials used were cylindrical pellets (6 mm diameter, 4 mm length) of pressureless sintered SiC containing a few percent alumina, and niobium foil (25 μm thickness) of 99.9 mass % purity. **Table 1** shows their physical properties. SiC/Nb/SiC couples were joined under vacuum below 1.33 mPa in a high frequency furnace equipped with a graphite susceptor (**Fig.1**). The applied joining pressure was 7.26 MPa. The joining

Table.1 Physical properties of Nb and SiC.

Matermal	Nb	SiC
Cofficient of thermal expansion (/K) (~1273K)	8.9×10^{-6}	4.7×10^{-6}
Bending strength (MPa)	------	430
Microhardness (kg/mm^2)	175	3000
Bulk density (g/cm^3)	8.57	3.13
Elastic modulus(MPa)	105000	392000
Melting point (K)	2468	2545

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temperature ranged from 1116 K to 1790 K. The joining time extended between 0.6~36 ks. The temperature and pressure cycle of this process is schematically illustrated in **Fig.2**. The phases occurring in the reaction zone were characterized by electron probe microanalysis(EPMA) and identified by X-ray diffraction (XRD).

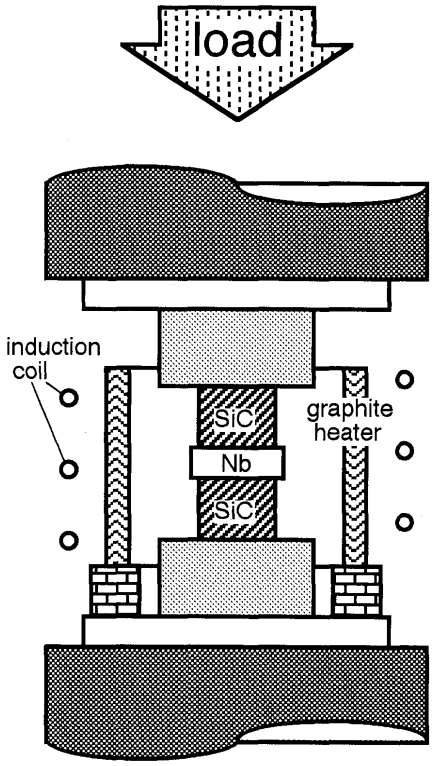


Fig.1 Schematic construstion for joining.

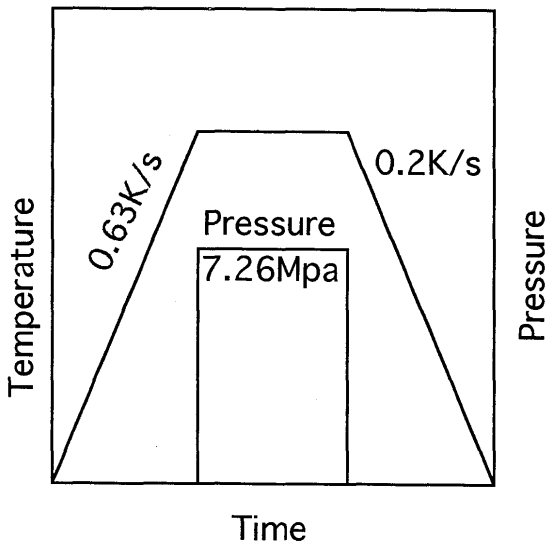
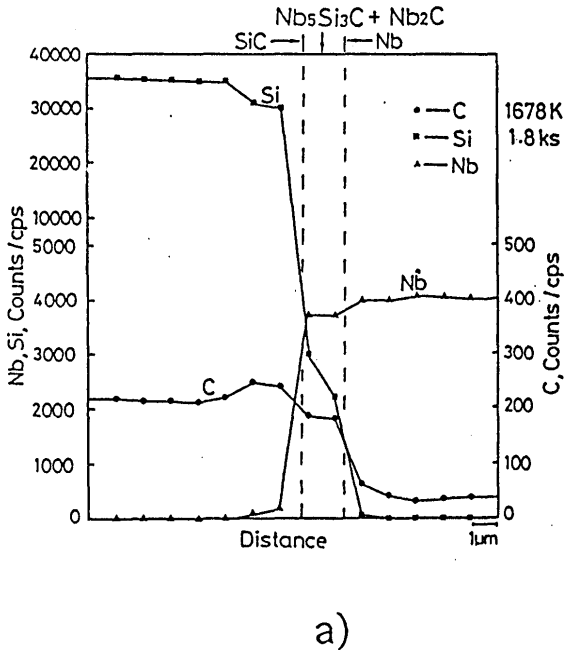


Fig.2 Temperature and pressure hysteresis during joining.

3. Results and discussion

3.1 Interfacial structure

Keeping the bonding time fixed at 1.8 ks and increasing the bonding temperature, interface roughness indicated an initiation of a reaction between SiC and Nb



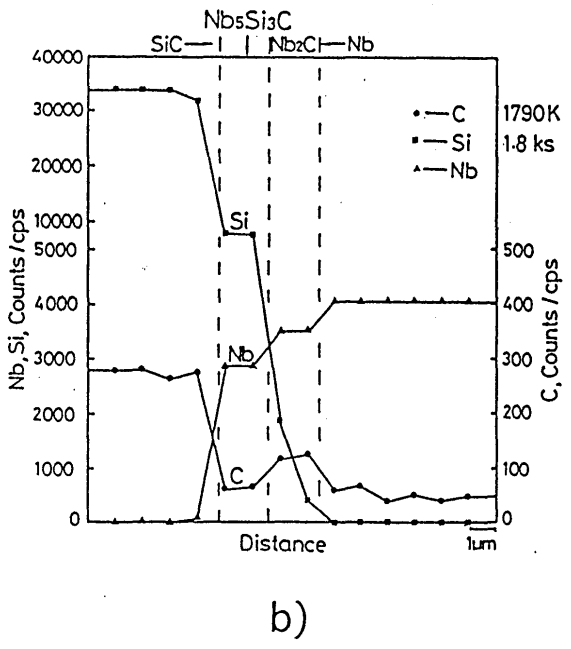


Fig.3 EPMA line analyses in SiC/Nb joint for 1.8ks, (a) at 1678 K, (b) at 1790 K.

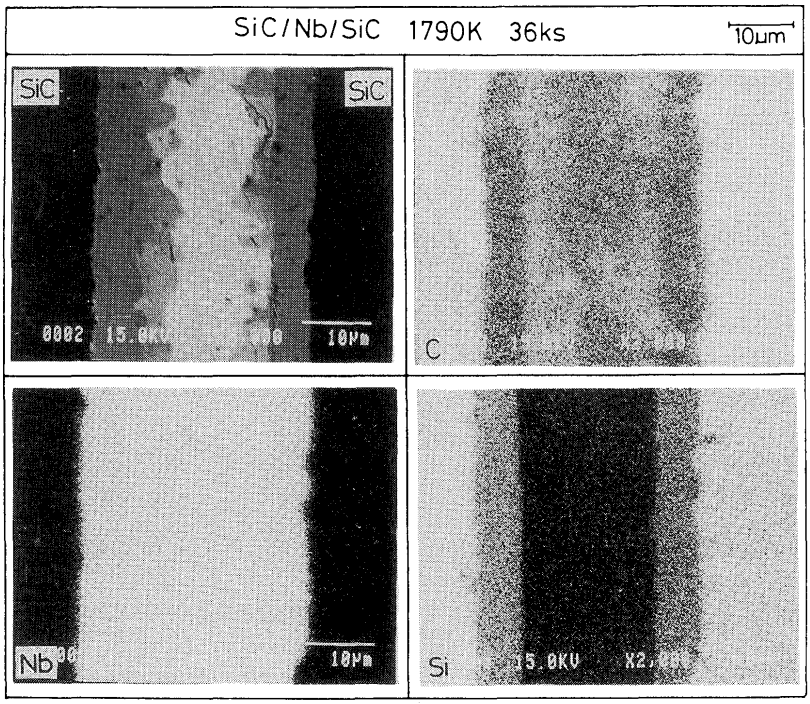


Fig.4 Microstructure and X-ray image analyses of elements in SiC joint with Nb at 1790 K for 36 ks.

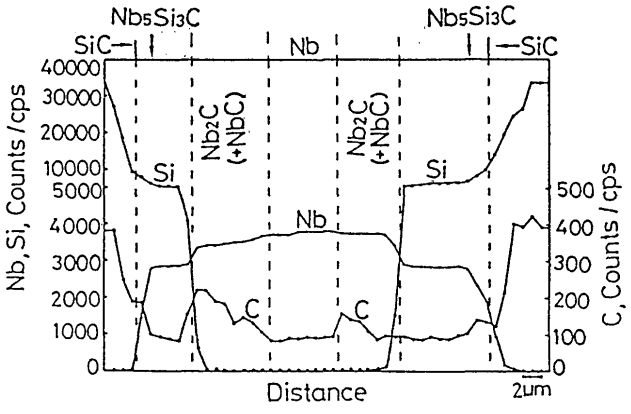


Fig.5 EPMA line analyses of Nb,Si and C in SiC/Nb/SiC joint at 1790 K for 36 ks.

at a temperature as low as 1116 K. At 1678 K a 1μm thin reaction layer could be detected. This can be clearly seen in **Fig.3a**, which shows the distribution of the elements C, Si and Nb across the reaction zone. At 1790 K (**Fig.3b**) the distribution of elements shows the formation of two layers. The layer next to SiC is high in silicon but depleted of carbon. The layer incontact with Nb shows higher carbon content, but little silicon. As will be discussed below, these phases are Nb_5Si_3C and Nb_2C, respectively.

The microstructure and distribution of elements after joining at 1790 K for 36 ks are shown in **Figs.4** and **5.** Elemental niobium remains at the central part. The reaction zone again consists of two layers. EPMA analysis shows the first layer, adjacent to SiC, to contain on the average 32.5 at% Si and 15.7 at% C. The actual values of measured scatter between 29.0 ~ 35.9 at% Si and 13.3 ~ 17.9 at% C. The resulting composition, Nb_5Si_3C, differs from the composition Nb_5Si_4C given for the ternary phase labled T in the Nb-Si-C phase diagram reported by Brukl[8], but corresponds to the composition given in a recent critical composition of the Nb-Si-C phase diagram[9]. X-ray analysis confirms the occurrence of the hexagonal Nb_5Si_3C Nowotny-type phasein that layer (**Fig.6**). Typical lattice parameters recorded are: a = 0.7540 nm and c = 0.5287 nm. **Fig.7** shows the microstructure and the elemental distribution for C, Si and Nb in that layer as revealed by mechanical polishing. The carbon rich phase is identified by XRD analysis to be NbC.

The second layer, adjacent to Nb in the central part of the SiC/Nb/SiC sandwich has an irregular morphology. EPMA yields an average composition of 70.0 at% Nb and 29.9 at% C. XRD identifies Nb_2C accompanied by small amount of NbC in that layer or adjacent to the Nb_5Si_3C containing layer.

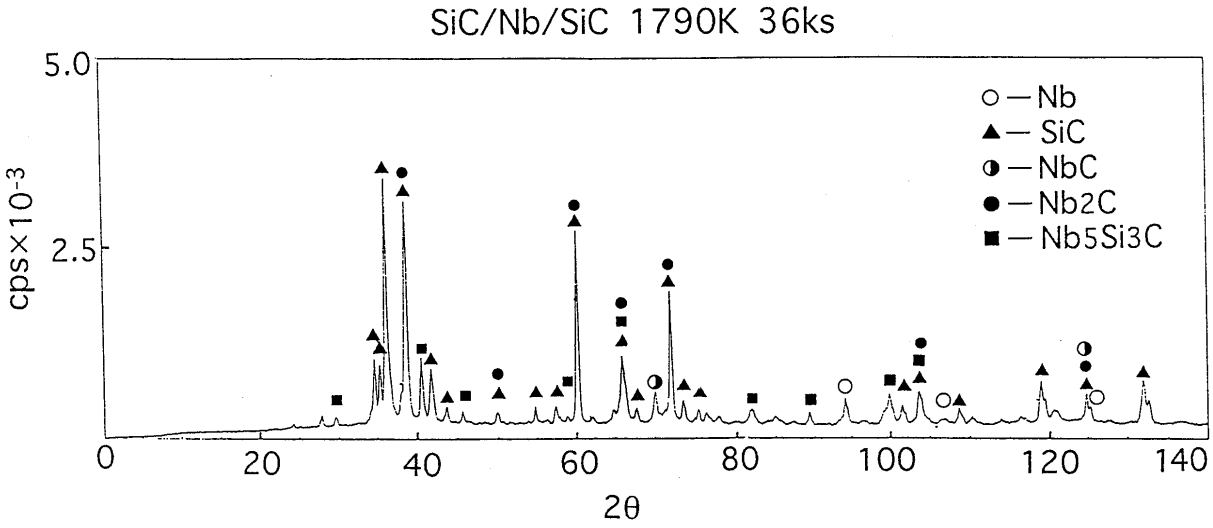


Fig.6 X-ray diffraction patterns of SiC/Nb/SiC interface at 1790 K for 36 ks.

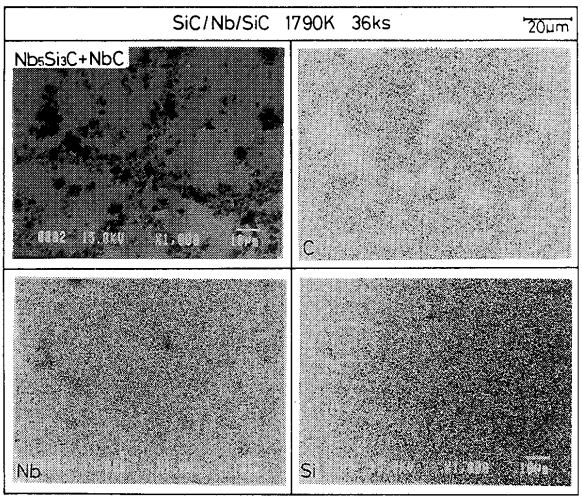


Fig.7 Microstructure and X-ray image analyses of elements in Nb_5Si_3C/Nb_2C polished interface at 1790 K for 36 ks.

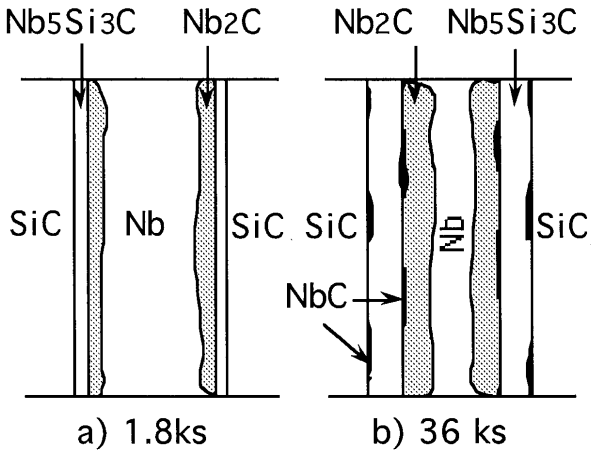


Fig.8 Schematic microstructures formed at 1790 K for 1.8 and 36 ks in SiC/Nb/SiC joint.

3.2 Reaction mechanism

As long as unreacted niobium remains at the central part, the reaction layers are mainly composed of Nb_5Si_3C and Nb_2C, respectively. NbC flakes are formed at the interface of the Nb_5Si_3C with SiC as well as at the interface of Nb_5Si_3C with Nb_2C. **Fig.8** shows schematically the resulting microstructure.

The phases at the interface between SiC and Nb couples are formed by the following reactions:

$$9Nb+3SiC=Nb_5Si_3C+2Nb_2C \quad (1)$$

$$7Nb+3SiC=Nb_5Si_3C+2Nb_2C \quad (2)$$

$$3SiC+5Nb=Nb_5Si_3C+2C \quad (3)$$

$$Nb_2C+2C=2NbC \quad (4)$$

Nb will react with SiC at the interface and forms Nb_5Si_3C. Carbon formed by eq. (3) will diffuse into Nb at the central part of the joint. The complete diffusion path is established at 1790 K after 36 ks, and expressed as SiC-NbC-Nb_5Si_3C-NbC-Nb_2C-Nb.

3.3 Growth of reaction layer

The thickness of Nb_5Si_3C and Nb_2C reaction layers are increased with the bonding temperature and with the reaction time. Their growth kinetics are studied from thickness measurements. The thickness of the reaction layer x increases linearly with the square root of the bonding time t :

$$x^2=k\cdot t \quad (5)$$

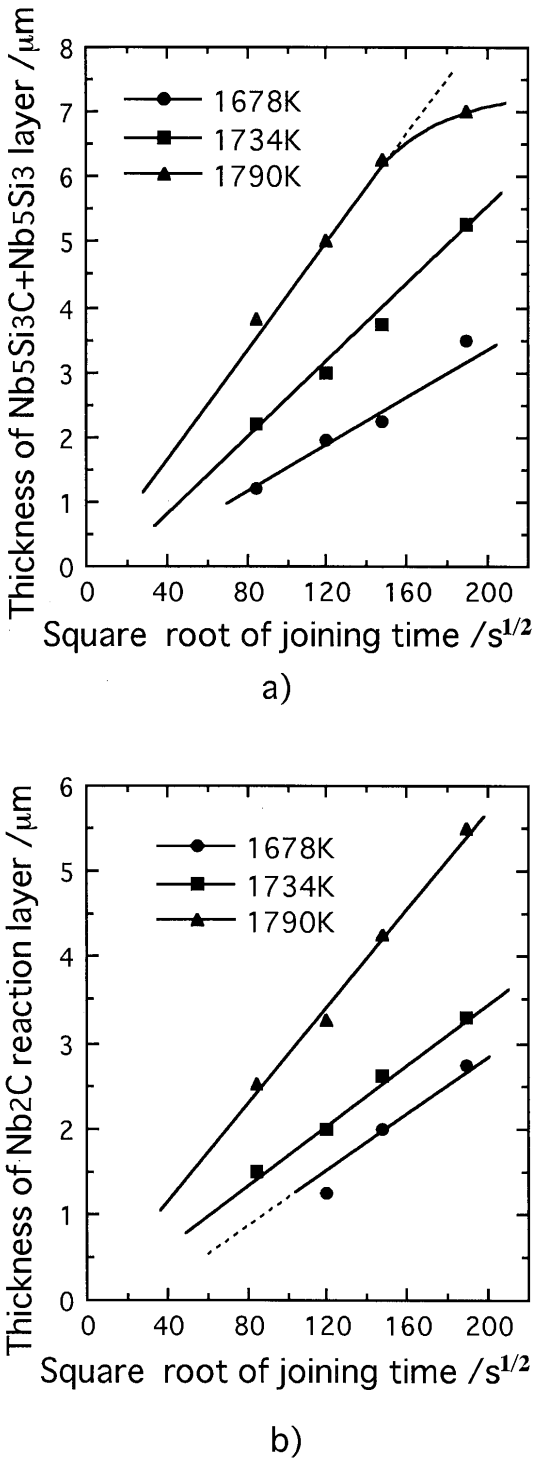


Fig.9 Relation between the reaction layer thickness and square root of bonding time, (a) Nb_5Si_3C phase, (b) Nb_2C phase.

where k is a material constant, as shown in **Fig.9**. The activation energy Q for growth of reaction layer is expressed as Arrhenius' law(5) in the bonding temperature range:

$$k=k_0 \cdot \exp(-Q/R \cdot T) \qquad (6)$$

Table 2 Values of Q and k_0 for reaction phases of SiC/Nb/SiC system

Composite	Q(kJ/mol)	$k_0(m^2/s)$	Refs.
Nb_5Si_3C	382	1.91×10^{-4}	Present work
Nb_2C	353	1.57×10^{-5}	Present work
Nb_5Si_3	452	-----	M.Naka
NbCx	49	-----	A.Joshi
NbCxSiy	83	-----	A.Joshi

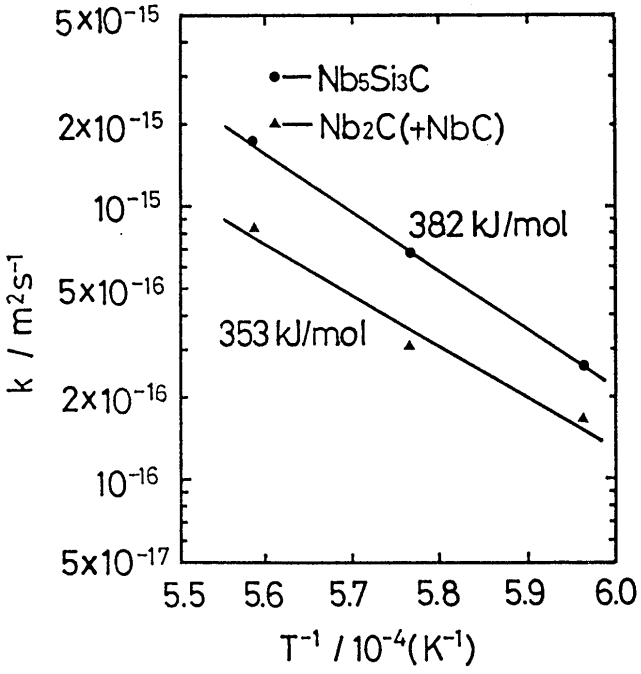


Fig.10 Rate constants k for Nb_5Si_3C and Nb_2C phase plotted against temperature.

where k_0 and Q are rate constant and activation energy for growth, respectively, as shown in **Fig.10**. The values of k_0 and Q are given in **Table 2**.

The activation energy of growth for NbCx and NbCxSiy reported by Joshi et al.[7] are quite lower than that of the present results. Since, the results by Joshi et al. have been measured in thin films of SiC/Nb system, defects such as dislocations in the film may accelerate the growth of phases.

The two reaction layers show different rate of growth as shown in **Figs.9** and **10**. The growth rate of Nb_5Si_3C is higher than that of Nb_2C. The k_0 factor of Nb_5Si_3C is larger than that of Nb_2C.Though Q factors of both phases are equal, k_0 factors affect the growth of both phases. The k_0 for the growth of Nb_2C is smaller

than that of Nb_5Si_3C, because C has to diffuse through Nb_5Si_3C. At 1790 K, for 36 ks, NbC formed at the interface between SiC and Nb_5Si_3C retards the growth of Nb_5Si_3C as shown in **Fig.9(a)**.

From the comparison of these results with those of Kurokawa and Nagasaki[4], the rate of reaction zone growth in SiC/Nb system is lower than SiC/Ti and SiC/Cr systems at same bonding conditions. This means that Nb possesses the lower reactivity against SiC compared with Ti and Cr.

The present results in **Fig.9** show that growth of reaction layers are mainly by diffusion of the elements, and also explain the importance of an accurate control of temperature and holding time.

4. Conclusions

The solid state bonding of SiC to SiC using Nb foil was conducted at the bonding conditions of 1116~1790 K, 0.6~36 ks and 7.26 MPa under a vacuum below 1.33 mPa. Interfacial reactions at the interface between SiC and Nb were studied using elemental analyses and X-ray difraction.

Reaction at the SiC/Nb interface was observed at temperatures of 1116 K and above. Upon bonding at 1790 K for 1.8 ks two layers form in the reaction zone. The layer next to SiC consists of Nb_5Si_3C. Adjacent to the remaining niobium predominantly Nb_2C is formed. At prolonged bonding time (36 ks) NbC phase appears at the interfaces SiC/Nb_5Si_3C and Nb_5Si_3C/Nb_2C. Thus the complete diffusion path appears to be SiC-NbC-Nb_5Si_3C-NbC-Nb_2C-Nb.

Growth of the reaction zones of Nb_5Si_3C as well as of Nb_2C (accompanied by NbC) follows Fick's law. The activation energies of growth for Nb_5Si_3C and Nb_2C are 382 and 353 kJ/mol, respectively.

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