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Interfacial Structure and Bond Strength of Solid State Bonded SiC/ Ni Joints†

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Abstract

SiC was bonded to SiC using Ni foil at temperatures ranging from 1223K to 1373K for 3.6 to 32.4ks and 30MPa in vacuum. SiC begins to join with Ni at temperatures above 1223K. At 1223K for 14.4ks, compounds δ-Ni_2Si, $Ni_{31}Si_{12}$ and Ni_3Si are formed in regular layers. Carbon cannot move to Ni, and remains as graphite in δ-Ni_2Si and $Ni_{31}Si_{12}$ layer zones as regular bands parallel to the original interface. The interface structure of the joint became SiC /δ-Ni_2Si+C(G) /$Ni_{31}Si_{12}$+C(G) /Ni_3Si /Ni. This microstructure represents a complete diffusion path which is correlated with the corresponding Ni-Si-C phase diagram. At the bonding condition of 1323K and a 32.4ks, Ni and Ni_3Si were completely consumed, and the joint showed the layer structure of SiC /δ-Ni_2Si+C(G)/ $Ni_{31}Si_{12}$+C(G)/ $Ni_{31}Si_{12}$. The bond strength of SiC/Ni/SiC joint depends on the thickness of the reaction layer zone between Ni and SiC, and shows a maximum value of 250MPa at 88 μm thickness.

KEY WORDS: (Bond Strength) (Interfacial Structure) (Ceramic) (Metal) (Diffusion) (SiC)(Ni)(Graphite) (Nickel Silicide)

1. Introduction

Ceramics get much attention as high hard and high temperature materials in many engineering fields. The mechanical properties of ceramics, however, frequently cause failure in applications as strucural components, because ceramics are brittle and have poor workability. Increasingly, engineering applications of ceramics require joining of the ceramics to ductile metals in order to overcome these problems.

SiC is well known as a high-temperature materials, and it was joined to many metals such as Ti[1], Cr[2], Nb[3],Ta[4] and V[5][6]. All these elements form carbides as reaction products during bonding.

Besides to these metals, SiC also was joined to Fe and Ni[7]-[11], elements which do not form carbides during bonding. Besides the considerable number of papers published about SiC/metal joints, the evolution of the microstructure of the interface and the strength of the joints have not been made clear in detail.

In the present study, SiC to SiC was bonded using Ni foil. The interfacial structure and elemental diffusion at the interface between SiC and Ni were correlated with the corresponding Ni-Si-C phase diagram. Farthermore, the bonding strength of the joint was investigated and is discussed in terms of the interfacial strucutre.

2. Experimental Procedure

The materials used were cylindrical SiC rods of 6 mm diameter and 4 mm thickness, containing a few percent of alumina as sintering aid (Kyocera,SiC211), and Ni foils of 200 μm thickness of 99.7 mass % purity (Nilaco,NI-313382). SiC /Ni /SiC couples were bonded in vacuum below 1.33mPa in a high frequency furnace equipped with a graphite tube. The applied bonding pressure was 30MPa. The bonding temperature ranged from 1223K to 1373K. The bonding time extended between 3.6 and 32.4ks. The phases occurring in the reaction zone were characterized by electron probe microanalysis (EPMA) , and identified by X-ray diffraction with CuKα diffraction. The strength of the joints was measured by shear testing using a cross-head speed of 1.7×10^{-5} m /s .

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3. Results and Discussion

3.1 Interfacial structure

SiC was solid state bonded to SiC using Ni foil. Interfacial microstructure and bond strength were studied at temperatures ranging between 1123K and 1323K for the bonding time extending from 3.6ks to 32.4ks.

Interfacial microstructure and quantitative analysis by EPMA for a joint bonded at 1223K for 14.4ks are shown in Fig. 1 and Table 1, respectively. Upon Ni reacting with SiC, Ni diffuses to SiC and forms $Ni_{31}Si_{12}$ and δ-Ni_2Si adjacent to SiC. At the same time Si diffuses into Ni and fomes Ni_3Si adjacent to Ni. On the other hand, carbon having as graphite a chemical activity of one can not move to Ni side.

Therefore, carbon remains in thin graphite layers in both δ-Ni_2Si and $Ni_{31}Si_{12}$ zones parallel to the original interface (Figs.1 a and b). The chemical composition analyzed for δ-Ni_2Si and $Ni_{31}Si_{12}$ zones conttaining graphite are given in Table 1. The white parts in the $Ni_{31}Si_{12}$+(G) zone and δ-N_2Si+(G) zone are $Ni_{31}Si_{12}$ and δ-N_2Si grains, respectively. The carbon found by EPMA analysis (points 2 and 4 in the Table 1) in the white $Ni_{31}Si_{12}$ and δ-N_2Si grains free of graphite may originate from graphite zone. Thus it is concluded, that the interface structure of the joint developed into layered SiC/ δ-N_2Si+(G)/ $Ni_{31}Si_{12}$+(G)/ Ni_3Si / Ni zones. The reaction layer zones increase in thickness with increasing the bonding time from 14.4ks to 28.8ks.

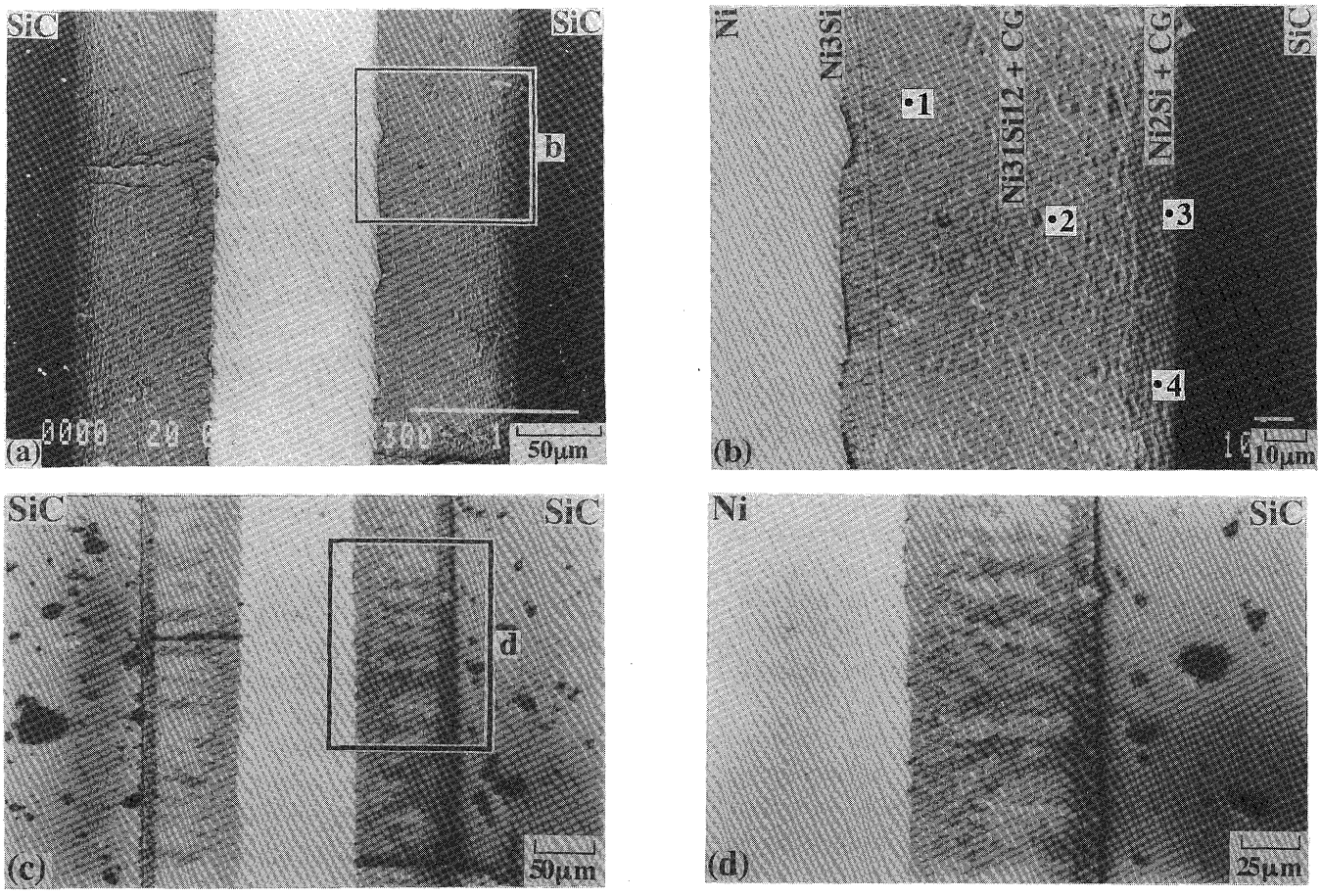


Fig. 1 Microphotograph of SiC/Ni/SiC joint bonded at 1223 K for 14.4ks.

Figs.1c and d shows optical microphotographs for a chemically polished specimen bonded at 1223K for 14.4ks. Fig.1d shows a very thin Ni_3Si layer adjacent to the Ni central part. The layer next to Ni_3Si represents $Ni_{31}Si_{12}$ containing graphite which preferred to grow perpendicular to the interface. The preferred growth orientation for $Ni_{31}Si_{12}$ is [7 7 $\bar{14}$ 16] perpendicular to the (11$\bar{2}$5) plan, which was deduced from the X-ray diffraction data. On the optical micrograph another layred zone, which was identified as δ-Ni_2Si+(G) by EPMA quantitative analysis, can be seen between $Ni_{31}Si_{12}$+(G) and SiC. In conclusion, the complete diffusion path between SiC and Ni following the sequence of SiC/ C(G)/ δ-Ni_2Si/ $Ni_{31}Si_{12}$/ Ni_3Si/ Ni was established at 1223K after 14.4ks. This path is shown by the broken line on the corresponding Si-Ni-C ternary phase diagram given in Fig.2.

Table. 1 Quantitative analysis of SiC/Ni/SiC joint bonded at 1223 K for 14.4ks.

Composition (at%)				Reaction Phases
No.	Ni	Si	C	
1	43.8	18.4	37.8	Ni31Si12 + CG
2	64.6	27.9	(7.5)	Ni31Si12
3	42.1	23.4	34.5	Ni2Si + CG
4	60.4	33.3	(6.3)	Ni2Si

Table. 2 Quantitative analysis of SiC/Ni/SiC joint bonded at 1323 K for 32.4 ks.

Composition (at%)				Reaction phases
No.	Ni	Si	C	
1	45.3	17.5	37.2	Ni31Si12 + Graphite
2	18.0	8.1	73.9	
3	57.4	39.3	13.3	Ni2Si + Graphite
4	35.2	17.4	47.4	
5	29.3	15.2	55.5	

The interfacial microstructure and quantitative analysis by EPMA for a joint bonded at 1323K for 32.4ks is shown in Fig.3 and Table 2, respectively. At the higher bonding temperature and longer bonding time, silicon still continues to diffuse from SiC, and Ni_3Si and Ni at the central part of joint were completely consumed, and changed to $Ni_{31}Si_{12}$ phase. Accordingly, the interface structure of this joint contains a sequence of SiC/ δ-N_2Si+(G)/ $Ni_{31}Si_{12}$+(G)/ $Ni_{31}Si_{12}$. Fig.4 shows X-ray diffraction patterns for different positions obtained by slight polishing from the central part of the joint to the SiC side (Figs.4 A, B and C). The intense peak (11$\bar{2}$5) of $Ni_{31}Si_{12}$ phase was obtained by polishing to B surface. This suggests that the preferred growth direction of $Ni_{31}Si_{12}$ phase was the [7 7 $\bar{14}$ 16] direction. The results of diffraction patterns of A and C agree with the quantitative analysis data shown in Table 2.

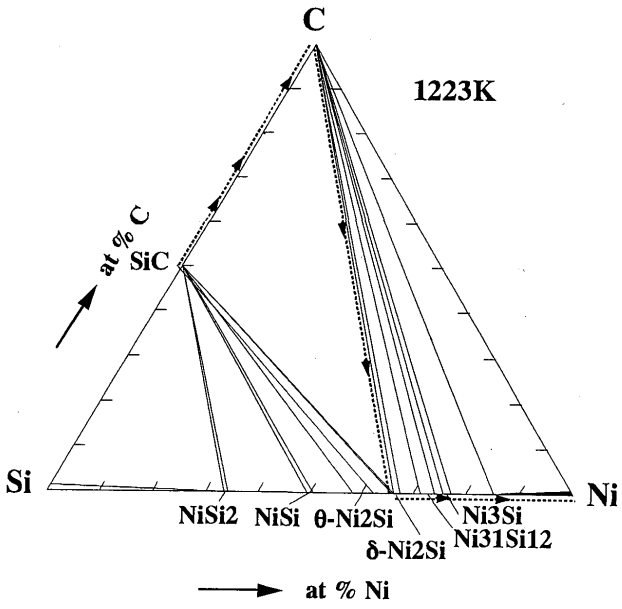


Fig. 2 Ni-Si-C ternary phase diagram and diffusion path in SiC/Ni/SiC system.

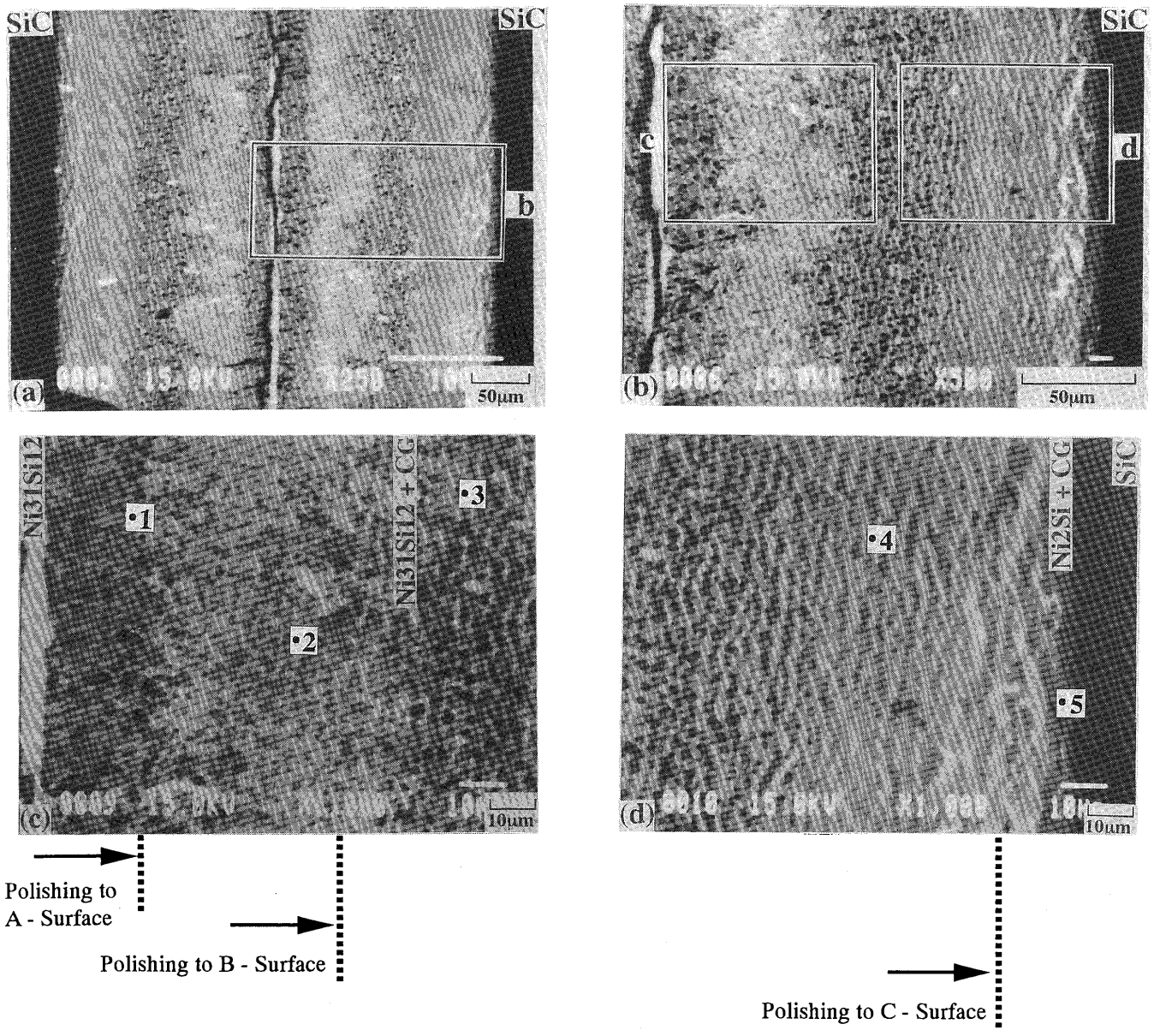
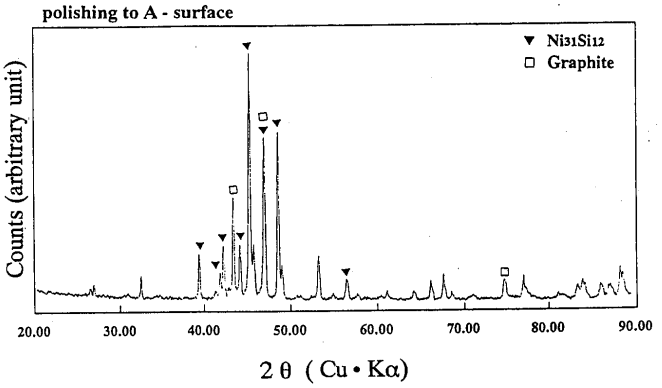
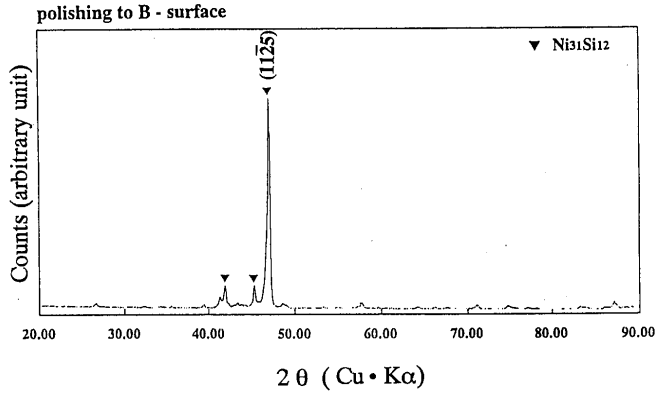


Fig. 3 Microphotograph of SiC/Ni/SiC joint bonded at 1323 K for 32.4 ks.





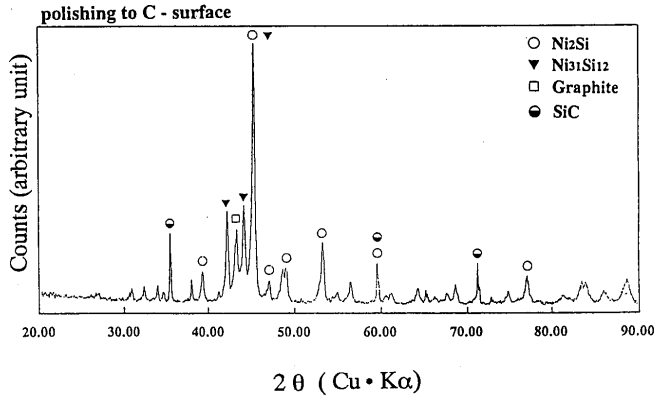


Fig. 4 X-ray diffraction pattern for different positions in joint bonded at 1323K for 32.4ks obtained by light polishing from central part of joint to SiC side.

3.2 Bond Strength

The bond strength of the SiC/Ni/SiC joints bonded at various bonding temperatures and bonding times was investigated as shown in Fig. 5. This figure shows the strength variation with the total reaction layer thickness of the SiC/Ni interfacial zones. The bond strength of the joint varies with the thickness of the reaction layer zone between SiC and Ni. The bond strength shows a maximum of 250MPa at a reaction zone thickness of 88μm, and the strength is markedly decreased in a reaction layer of 120μm.

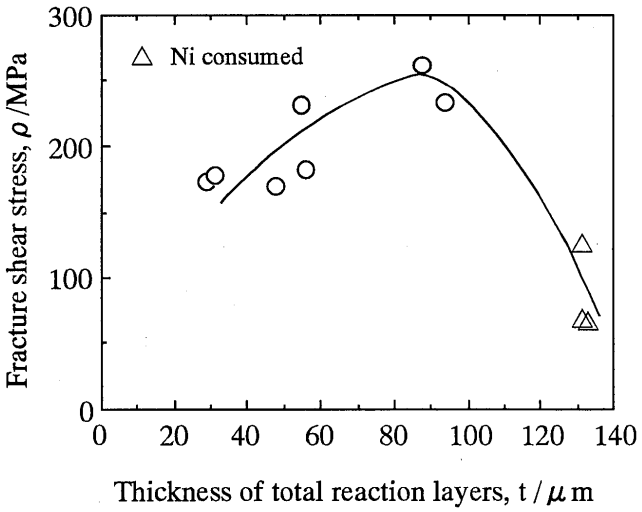


Fig. 5 The variation of SiC/Ni interface strength with thickness of total reaction layers.

The microphotographs of fracture surfaces and quantitative analysis by EPMA for the joint bonded at 1223 K for 14.4 ks are shown in Fig.6 and Table 3, respectively. This joint had a reaction layer zone of 31μm thickness and a bond strenght of 178MPa. XRD shows the presence of $Ni_{31}Si_{12}$ and graphite at the fractured surface (Fig.7). Again, $Ni_{31}Si_{12}$ seems to have grown preferentially oriented along (11$\bar{2}$5). Compositions of point 2 (light gray) and 4 (white) represents the prefentially oriented growtn $Ni_{31}Si_{12}$ phase, while the compositions of point 3 (gray) and 5 (black) represents graphite and SiC, respectively. These results indicate that the fracture took place in the prefentially oriented grown $Ni_{31}Si_{12}$ phase. The relatively high strength of the joint suggests that the grain boundary of the preferred growth phase served to protect against the crack propagation.

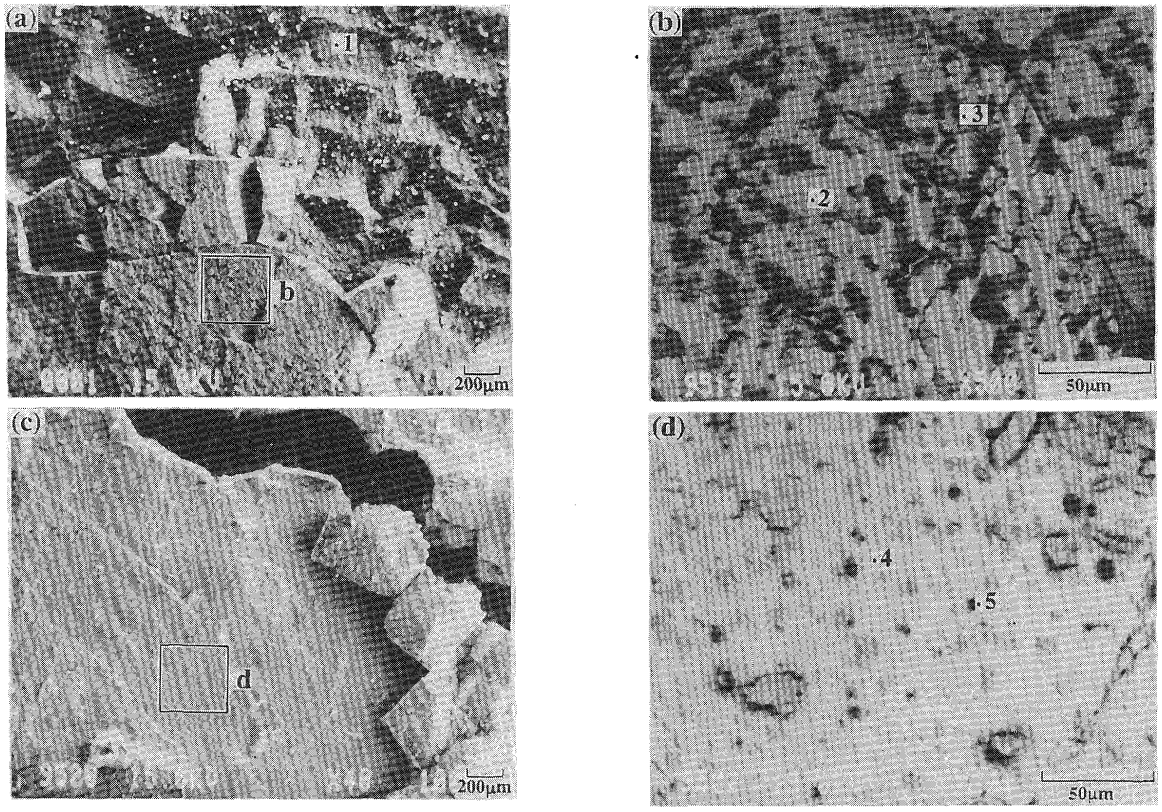


Fig. 6 Microphotograph of fracture surface for SiC/Ni/SiC joint bonded at 1223 K for 14.4 ks. (a) SiC side (b) The magnification of (a), (c) Ni side (d)The magnification of (c).

Table. 3 Quantitative analysis of fracture surface bonded at 1223 K for 14.4 ks

Composition (at%)				Reaction pheses
No.	Ni	Si	C	
1	0.0	45.6	54.4	SiC
2	66.5	27.4	6.1	Ni31Si12 + CG
3	22.2	9.1	68.7	Graphite
4	69.8	27.2	3.0	Ni31Si12
5	0.0	46.5	53.5	SiC

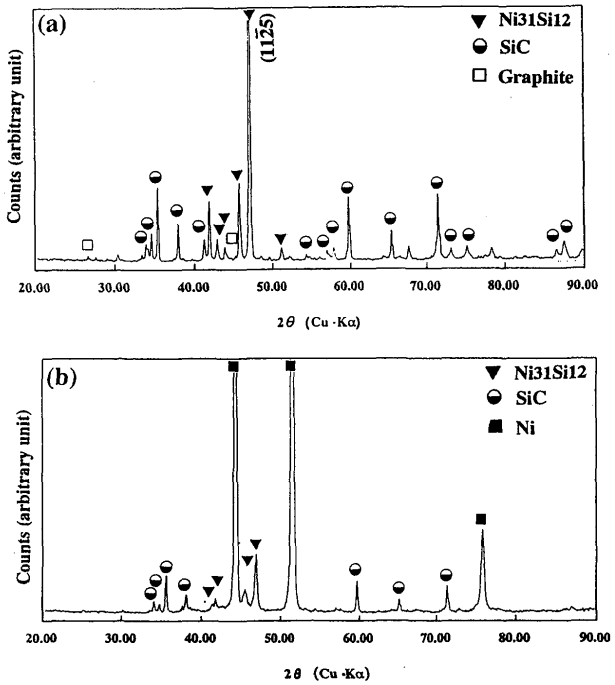


Fig. 7 X-ray diffraction pattern of the fracture surface of SiC/Ni/SiC joint bonded at 1223 K for 14.4ks. (a) SiC side , (b) Ni side

Next, the microphotographs of fracture surfaces and quantitative analysis by EPMA for the joint bonded at 1323K for 32.4ks are shown in Fig.8 and Table 4, respectively. This diffusion couple shows a value of 74MPa at the thickness 132μm of the reaction layer. Composition at point 1 (white) represents $Ni_{31}Si_{12}$ single phase, while the composition for point 2 (black) and 3 (gray) represents graphite and δ-Ni_2Si +(G) respectively. The crack in this joint which showed the low strength, propagated from δ-Ni_2Si +(G) zone of SiC side to $Ni_{31}Si_{12}$ single phase zone which both zone are mechanically brittle.

Table. 4 Quantitative analysis of fracture surface bonded at 1323 K for 32.4 ks.

Composition (at%)				Reactiom pheses
No.	Ni	Si	C	
1	59.2	35.3	5.5	Ni31Si12
2	4.1	3.2	92.5	Graphite
3	37.5	19.2	42.5	Ni2Si + Cg

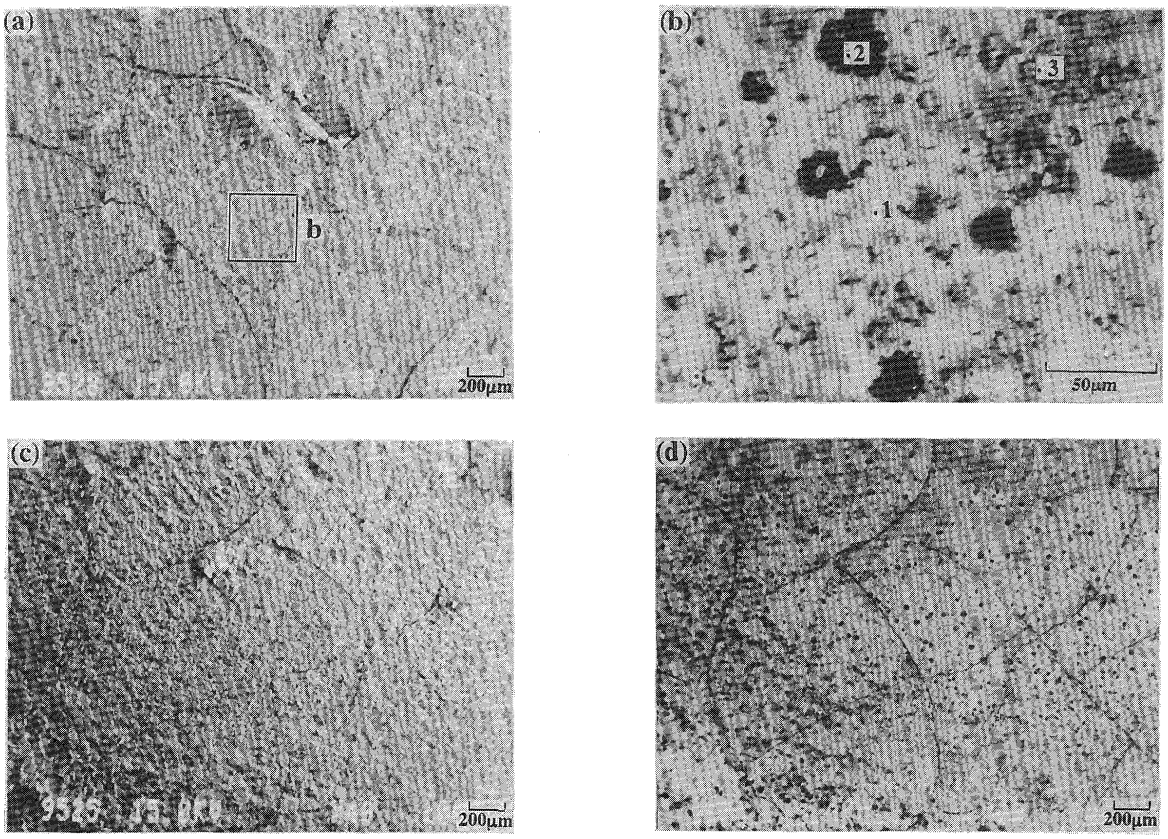


Fig. 8 Microphotograph of fracture surface for SiC/Ni/SiC joint bonded at 1323 K for 32.4 ks.
(a) Fracture surface of reaction layer , (b) The magnification of (a)
(c) The oppsite Surface of (a) , (d) The backscattered electron image of (c).

4. Conclusions

The solid state bonding of SiC to SiC using Ni foil was conducted at the bonding conditions of 1223 to 1323K, 3.6 to 32.4ks and 30MPa in vacuum below 1.33mPa. Interfacial microstructures and bond strengths were investigated using electron probe microanalyser, X-ray diffractometer and fracture shear testing. At the bonding condition of 1233K for 14.4ks, Si diffuses into Ni, and forms Ni_3Si adjacent to Ni. Ni also diffuses to SiC and forms $Ni_{31}Si_{12}$ adjacent to Ni_3Si and δ-Ni_2Si adjacent to SiC, respectively. C cann't diffuse to Ni, and remains as graphite layers in δ-Ni_2Si and $Ni_{31}Si_{12}$ zones parallel to the original interface. The interface structure of SiC/ δ-N_2Si+(G)/ $Ni_{31}Si_{12}$+(G)/ Ni_3Si / Ni represents the diffusion path connecting SiC to Ni on the phase diagram of Ni-Si-C. At the bonding condition of 1323K for 32.4ks Ni and Ni_3Si are completely consumed, and the interface strucure of the joint changes to SiC/ δ-N_2Si+(G)/ $Ni_{31}Si_{12}$+(G)/ $Ni_{31}Si_{12}$. The strength of the joint depends on the totall thickness of the reaction layers between SiC and Ni. The strength of the joint bonded at 1273K for 14.4ks having a total reaction zone thickness of 88μm shows a strength maximum of 250MPa .

REFERENCES

1) M. Naka , J. C. Feng, andJ. C. Schuster, Met. and Mater.Trans.A, 28A(1997), 1385- 1390.
2) J.C. Feng, M. Naka and J. C. Schuster, J. Japan Inst.Metals, 61(1997),636-642.
3) M. Naka , J. C. Feng,J. Japan Inst.Metals, 59(1997),79-83.
4) J.C. Feng, M. Naka and J. C. Schuster, J. Japan Inst.Metals, 61(1997),456-461.
5) T.Fukai, M. Naka and J. C. Schuster, in Interface Science and Interconnection, Proc.JIMIS-8 Jap.INst.Metals, Japan, 1996 p 287-290.
6) T.Fukai, M. Naka and J. C. Schuster,Trans,Joining and Weld.Inst.26(1997),93-98.
7) W. Jeitschko, and H. Nowotny, Monatsh. Chem.,98(1967), 328-337.
8) J. C. Schuster, Structural Ceramics Joining II, Ceramic Transactions, 35(1993), 43-57.
9) M. R. Jackson, R.L.Mehan, and A.M. Davies, and E.L.Hall, Met. Trans. A14(1983), 355-364.
10) R.C.J.Schiepers, F.J.J.van Loo and G.de With, J. Amer. Ceram.Soc.71 (1988) C284-C287.
11) K.Kurokawa and R.Nagasaki, Proc.Int.Symp Science of Sintering (IISS), Tokyo. Elsevier Sci., New York, USA,1988 p1397-1402.