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Interface Layer Structure of Solid State Bonded SiC/TiAl Joint†

Akinori Inoue*, Hiroshi Miyake*, Masaaki Naka,Toshiya Shibayanagi*** and Masakatsu Maeda******

Abstract

Solid state bonding of SiC to TiAl has been performed at temperatures from 1473 K to 1573 K. The microstructure at the SiC / TiAl interface and the growth mechanisms were analyzed by EPMA analysis and X-ray diffractometry. The interface structures were the same at all bonding temperatures. The reaction layers were divided into two parts. The first one was a Ti carbide based layer, and the other was a Ti silicide based layer. Both layers are mixed layers composed of one or two carbides and silicides including Ti_3AlC ternary compound. At the reaction layer side in TiAl, a Ti depleted zone and an Al rich TiA1 1ayer were formed since Ti in TiAl reacted and diffused toward the SiC side. The reaction layer formed at SiC / TiAl joint was determined as,

SiC / TiC / TiC+Ti_3AlC / Ti_5Si_3+$TiSi_2$+Ti_3AlC / TiAl (Al-rich) / TiAl

The reaction layers grow in proportion to the square root of the bonding time. The growth activation energy of the carbide layer containing TiC and Ti_3AlC, the silicide layer and the total reaction layer were 398 kJ/mol, 553 kJ/mol and 502 kJ/mol. The growth of the total reaction layer was dominated by the growth of the silicide, because the main part of thickness consisted of the silicide.

KEY WORDS: (Silicon carbide)(Intermetallics)(TiAl)(Solid state bonding)(Titanium Carbide) (Titanium Silicide)

1. Introduction

Joining of ceramics to metals has been playing a quite important role in extending the applications of advanced ceramics in many kinds of industries. Among the engineering ceramics, SiC has been attracting much attention, since it is well known to have excellent hardness and resistance to high temperature. The joining of SiC to high temperature metallic materials such as Ti, Nb, Cr and V has been investigated by means of the solid state diffusion bonding technique[1-4].

In order to raise the service temperature as well as to increase the reliability of the joints, strong interest has been focusing on some new advanced materials such as intermetallics that have excellent high temperature properties.

TiAl is one of the promising high temperature intermetallics, and extensive research work has been performed to apply this compound as a light structural material to be used both at ambient temperature and at high temperature[5]. For this reason, TiAl is now a candidate material as a counterpart of the joining of SiC, although no information has been obtained so far for the interfacial reaction process between the two materials at elevated temperature.

The present work is aiming to identify the reaction products and essential processes of the interfacial reaction between SiC and TiAl during solid state diffusion bonding.

2. Experimental procedure

The materials used were cylindrical SiC rods of 6mm in diameter and 4mm in length, containing a few percent of alumina as a sintering aid, and TiAl foils of 100 μ m thickness. γ -TiAl ingots containing 53.0at% Al were prepared by the arc melting method. After homogenizing at 1273K for 432ks, foils of 500 μ m in thickness were sliced off and ground to 100 μ m in thickness followed by the final polishing using 0.3 μ m Al_2O_3 powders.

SiC / TiAl / SiC couples were bonded in vacuum below 1.0mPa in a high frequency furnace equipped with a graphite tube as shown in **Fig.1**.

Bonding temperature was measured with an infrared radiation thermometer from the outside of the chamber. The applied bonding pressure was 200MPa. The bonding temperatures were 1523K,1573K and 1623K. The bonding time were 7.2ks, 28.8ks and 115.2ks.The phases existing in the reaction zone were identified by both

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EPMA and X-ray diffraction technique using Cu Kα.

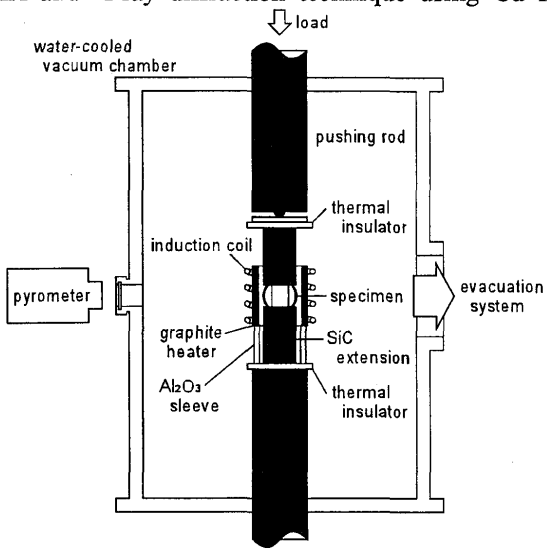

Fig.1 Schematic drawing of the bonding apparatus.

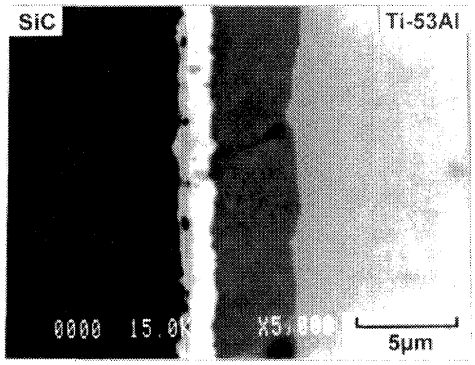

Fig.2 Back-scattered electron image of SiC/TiAl interface bonded at 1473K for 14.4ks.

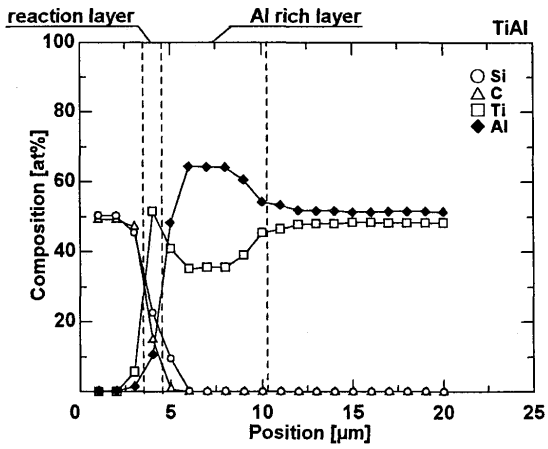

Fig.3 Elemental line analysis of SiC / TiAl interface bonded at 1473K for 14.4ks.

3. Results and discussion

3.1 Interfacial phase reaction

Figure 2 shows the results of SEM observation of the interfacial region of the joint bonded at 1473K for 14.4ks. The line analysis of composition by EPMA method was also performed for the same area in Fig.2 and the result is shown in Fig.3. In Fig.2, the region on the left side with dark contrast is SiC, and TiAl is observed as a brighter region existing in the right side. In between these starting materials, two layers with different contrast appear. As explained in the later section, the layer adjacent to SiC is TiC. The reaction layer next to the TiAl is composed of two mixed phases. The first one contains titanium-based silicides such as Ti_5Si_3 and TiSi. The second one is composed of the mixed phases of Ti_5Si_3, $TiSi_2$ and a ternary phase of Ti_3AlC. The ternary phase in the reaction layer possesses granular shapes of sizes from 0.5 to 0.7 μ m in diameter. In the TiAl region adjacent to the reaction layer, there is a zone with darker contrast than that of TiAl. According to the line analysis of composition as shown in Fig.3, the zone contains a

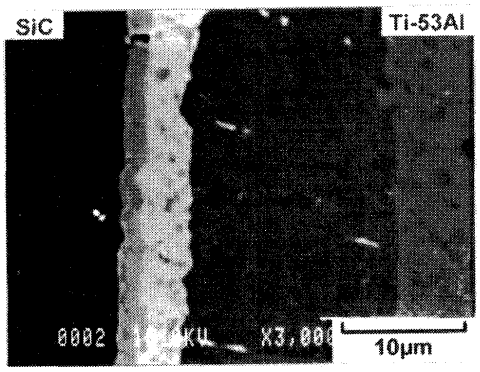

Fig.4 Back-scattered electron image of SiC/TiAl interface bonded at 1573K for 14.4ks.

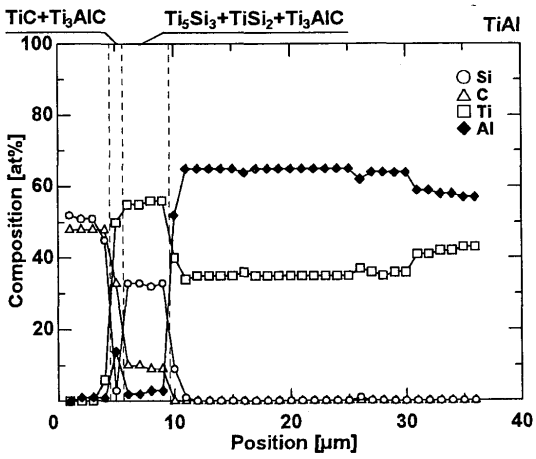

Fig.5 Elemental line analysis of SiC/TiAl interface bonded at 1573K for 14.4ks.

smaller amount of Ti (the maximum amount of Al was 64 at%) than the initial composition of TiAl.

This result means that Ti in TiAl reacted preferentially with SiC and was diffusing into the reaction layers.

Besides the formation of new phases between SiC and TiAl during diffusion bonding, formation of reaction products yielded the wavy interfaces as seen in Fig.2.

The interfacial microstructure of the joint bonded at the higher temperature of 1573K was then analyzed by means of SEM observation and EPMA techniques, and the results are shown in **Figs. 4 and 5,** respectively. The bonding time is maintained at 14.4ks. Different phase reactions occurred in the region adjacent to the SiC, and the new region was estimated to consist of mixed phases of TiC and Ti_3AlC. A TiC layer existed between the SiC and the mixed phases layer. The TiC layer was observed more clearly as the bonding time increased to 140ks, which will be explained later. The reaction layer on the TiAl side contains about 10 mass% of Al, and it should be a mixture of Ti_5Si_3, $TiSi_2$ and Ti_3AlC phases. There is a granular phase in this region, and the phase is mainly

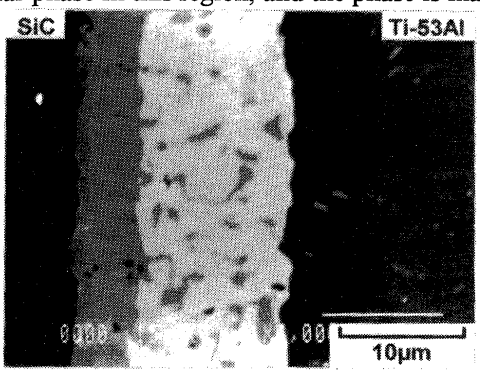

Fig.6 Back-scattered electron image of SiC/TiAl interface bonded at 1573K for 130ks.

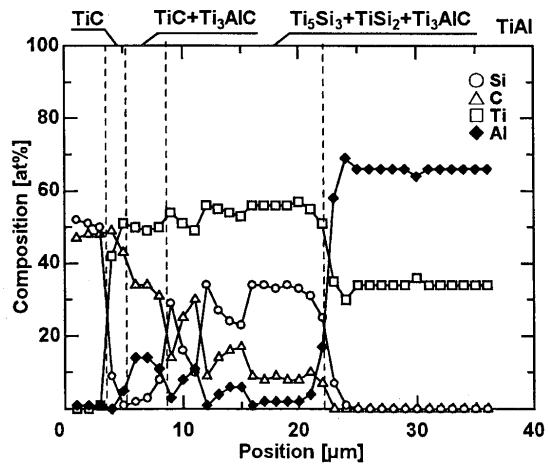

Fig.7 Elemental line analysis of SiC/TiAl interface bonded at 1573K for 130ks.

Ti_3AlC granules having diameters ranging from 0.6 to 1 um. The TiAl showed a Ti depleted zone with darker contrast having 64 at% of Al content, which is adjacent to the reaction layer.

Figures 6 and 7 show the results of SEM observation and EPMA analysis of the interfacial region of the joint bonded at 1573K for the prolonged time of 130ks, respectively. Beside SiC there is a reaction layer having a composition corresponding to TiC, and the next layer adjacent to the TiC layer seems to be composed of the mixed phases of TiC and Ti_3AlC containing some 10 mass% of Al. The Ti_3AlC is a granular phase showing slightly darker contrast. Beside the TiAl a reaction layer of 10um in thickness is observed, having the composition corresponding to the mixed phases of Ti_5Si_3, $TiSi_2$ and Ti_3AlC. The Ti_3AlC phase in this reaction layer has granular form and corresponds to a C rich region in the composition profile. The Al content in the TiAl is no longer the initial one, and tends to increase towards the reaction region. About 70at% of Al is measured in the region just beside the reaction layer. Phase identification of these reaction products was then performed carefully by means of the X-ray diffraction technique, and the results will be explained in the following section.

Figure 8 shows the results of X-ray diffraction analysis for the joint bonded at 1573K for 130ks. The patterns were taken from the SiC side in the interfacial region of the joint. The peaks marked with ▼ correspond to the SiC (cubic structure with the measured values of lattice constant a=4.367Å, a=4.3589Å form the literature[6]). The peaks marked with ○ correspond to the TiC (cubic structure, a=4.339 Å, the literature value[7] is a=4.3274 Å). The peaks marked with □ correspond to the Ti_5Si_3 (hexagonal structure, a=7.422 Å, c=5.148Å, the literature values[8] are a=7.444 Å, c=5.143Å). The peaks marked with ■ correspond to the $TiSi_2$ (monoclinic structure, a=8.262 Å, b=8.860 Å, c=4.7778Å, the literature values[8] are a=8.2687 Å, b=8.5534Å, c=4.7983Å). There were also observed $TiSi_2$ and TiC as major components along with SiC in the measured region of the joint.

Figure 9 represents the XRD pattern taken from the interfacial region close to the TiAl. The peaks of Ti_5Si_3, SiC and $TiSi_2$ are observed. TiC seems not to exist in this region, but the region row contains Ti_3AlC marked with ◎ (cubic structure, a=4145Å, the literature value[9] is a=4.156 Å). The $TiSi_2$ has very strong intensity.

Summarizing the results of SEM observation, EPMA analysis and XRD analysis explained in the figures from

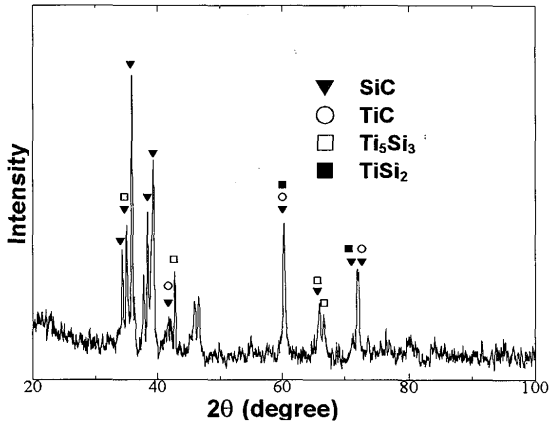

Fig.8 X-ray diffraction pattern from SiC side of SiC/TiAl interface bonded at 1573K for 130ks.

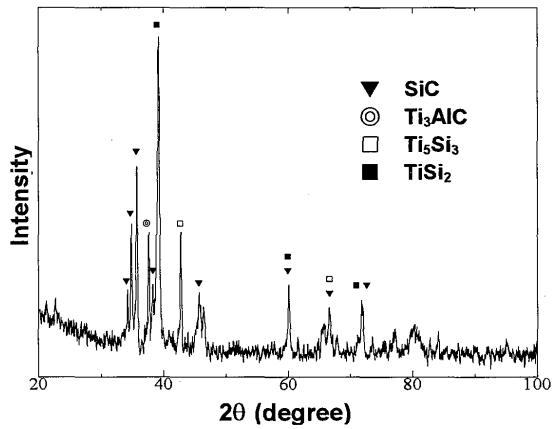

Fig.9 X-ray diffraction pattern from TiAl side of SiC/TiAl interface bonded at 1573K for 130ks.

6 to 9, the granular carbides existing in the layer of mixed titanium carbide phases is finally identified as Ti_3AlC. The matrix is TiC. Since the peak intensity of Ti_3AlC is strong, the granular phase in the Ti_5Si_3 layer adjacent to SiC is also Ti_3AlC. The Ti_3AlC still exists in the reaction layer of TiSi.

Consequently, from the phase identification explained so far, the interfacial phase sequence of SiC/TiAl joint bonded at 1573K for 130ks is concluded as follows;

SiC / TiC / TiC+Ti_3AlC / Ti_5Si_3+$TiSi_2$+ Ti_3AlC / TiAl(Al-rich) / TiAl

3.2 Growth kinetics

This section will discuss the growth mechanisms based on the results explained in the previous section. Generally the growth behavior of reaction phases between ceramics and metals is controlled by the diffusion process of the constituents in each reaction product. The diffusion controlled reaction process is described by the following equation;

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

where X is the thickness of reaction layer, k is a time constant and t is the bonding time.

The time constant k is given by the following equatin;

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

where k_0 is called growth factor, Q is the activation energy, R is the gas constant and T is the bonding temperature.

According to the equation 1, the thickness of reaction layer increases proportionately with the square root of the bonding time. Taking logarithms of both terms, the equation 2 is transformed as follows;

$$\ln k = \ln k_0 - Q/RT \qquad (3)$$

Utilizing the equation 3, the values Q and k_0 for each reaction layer observed in the present study were calculated in the following analysis. In the present work, the Q is called an "apparent activation energy" since it reflects many elementary processes such as diffusion and phase reaction.

Tite relationships between the thickness of each reaction layer and the bonding time at each bonding temperature were analyzed, and are represented in the figures from 10 to 12.

Thickness of the reaction layer composed of (TiC+Ti_3AlC) mixed phases changes as shown in **Fig. 10**. Each mark △, ■ and ○ represent the data taken at the bonding temperatures of 1473K, 1523K and 1573K, respectively. Except for the data taken for the specimens bonded for longer time at 1523K and 1573K, the thickness of the reaction layer increases linearly, indicating that the phase reaction is the diffusion controlled process. For the joints bonded for the longer time, the thickness of (TiC + Ti_3AlC) layer tended to be less than the expected value given by the square root rule of reaction kinetics as shown in **Fig.10**. This phenomenon could be attributed to some change in the diffusion process of each constituent caused by the alteration of the composition of TiAl adjacent to the reaction layer.

Figure 11 shows the growth kinetics of the (Ti_5Si_3+$TiSi_2$+ Ti_3AlC) mixed phases. Good linearity was observed for each data type and the slope increased as the bonding temperature was raised.

Figure 12 shows the change of total thickness of reaction layers, indicating that the data also show good linearity while the approximated lines start from the origin. Similar to the data shown in Fig.11, the slope

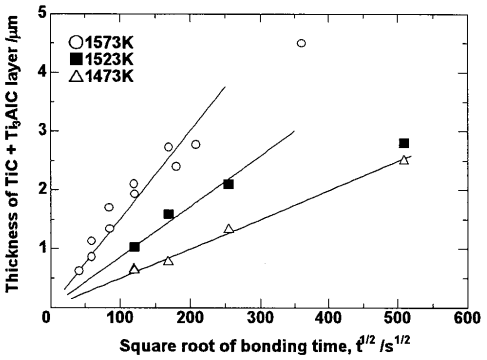

Fig.10 Thickness of TiC+Ti_3AlC layer plotted with a square root of the bonding time.

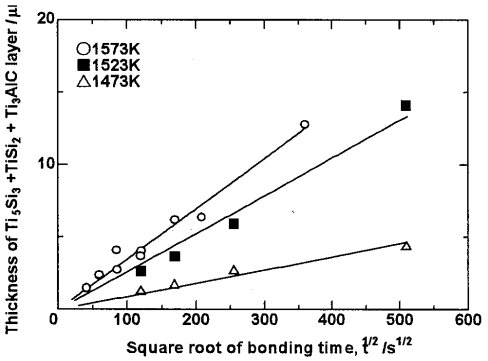

Fig.11 Thickness of Ti_5Si_3+$TiSi_2$+ Ti_3AlC layer plotted against the square root of the bonding time.

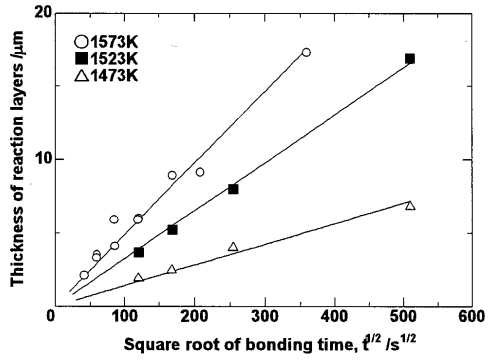

Fig.12 Total thickness of reaction layers plotted against the square root of the bonding time.

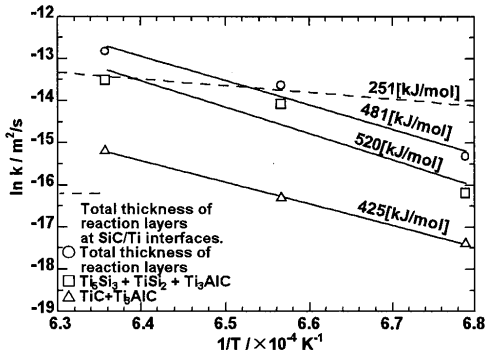

Fig.13 Reaction constant k plotted with ln(T) , where T is the bonding temperature.

increased with bonding temperature. As the thickness of the silicides layer is larger than that of the (TiC+Ti_3AlC) mixed layer, total thickness of the reaction layers is close to that of the layer of the silicides.

Figure 13 represents the Arrhenius plots for the data shown in the figures from 10 to 12. Note that the data which deviated from linearity are not plotted. The marks ○, □ and △ are the data for total thickness, mixed silicides layer and carbides layer, respectively. The apparent activation energy for each layer was calculated from each slope in this figure. The values Q were 502,553 and 398 kJ/mol. for the carbides layer, silicides layer and total reaction layers, respectively.

Naka et al[10] reported the activation energy of 267kJ/mol. for (Ti_5Si_3Cx + TiC) mixed phases in the SiC/Ti joint, as indicated by a dashed line in Fig.13. This value is significantly smaller than the value of 481kJ/mol. that was obtained for the total reaction layers in the present work. The difference is caused by the activity of Ti. In other words, TiAl is a more stable phase than pure Ti, therefore much more energy is required for the Ti in TiAl to react with SiC and to diffuse in to the adjacent region.

4. Conclusions

Solid state diffusion bonding of SiC to TiAl intermetallics has been performed at temperatures from 1473K to 1573K, and the interfacial reaction process was analyzed regarding the identification of phases and kinetics of phase reaction at the interface. The following results were obtained.

(1)Every specimen bonded at each temperature possesses the same interfacial phase sequence as,

SiC/TiC / TiC+Ti_3AlC / Ti_5Si_3+$TiSi_2$+Ti_3AlC / TiAl(Al-rich) / TiAl

Titanium based carbide layers exist at the SiC side, and a mixture of titanium based silicides is adjaceat to TiAl. Each reaction layer is composed of a ternary compound of Ti_3AlC, and other phases such as silicide or carbide. A preferential diffusion of titanium from TiAl toward the SiC side was concluded to yield a Ti depleted zone between the reaction layer and TiAl.

(2)Each reaction layer grew in proportion to the square root of the bonding time. The apparent activation energy for the growth of the carbide layer consisting TiC and (TiC+Ti_3AlC) mixed region was calculated

to be 425kJ/mol. The silicide layer of (Ti_5Si_3+$TiSi_2$+Ti_3AlC) mixed phases region was calculated to have the apparent activation energy of 520kJ/mol for growth. The activation energy of total phase reaction was 481kJ/mol. The silicide layer was the major part of the reaction zone, and its growth process was concluded to control the phase reaction at the interface between SiC and TiAl.

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