Effect of Sn Addition to Cu-Ti Filler Metals on Microstructure and Strength of SiC/SiC Joint †

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Abstract

SiC was brazed to SiC in vacuum using two series of Cu-16.5 at%Sn-X at%Ti and Cu-X at%Sn-21 at%Ti alloys. At a constant Sn content of 16.5 at% the strength of the SiC/SiC joint shows a maximum at 5.6 at%Ti , and decreases with the further content of Ti in the alloys. Two factors operates to yield the maximum strength of the SiC/SiC joint. The fine Ti_5Si_3 precipitates distribute densely at the SiC/the alloy interface, and α-Cu solid solution containing Sn and δ-Cu-Sn intermetallic yield a two phase eutectoid structure. The excess amounts of Sn and Ti in the alloys form a brittle intermetallic of $CuSn_3Ti_5$ in the joining layer of SiC/SiC, and affect the strength of the joint.

KEY WORDS: (Brazing) (Ceramic-metal Joining) (SiC) (Tin) (Copper) (Titanium) (Interfacial Strucutre)

1. Introduction

Silicon carbide is a candidate material for structural ceramics. Joining of ceramics enlarges possible engineering applications [1]. It is known that copper base alloys containing Ti easily wet SiC, and are applicable to the brazing filler metal [2]. The brazing of SiC has been tried by using Ag-Cu-Ti [3] and Cu-Ti [4,5] alloys. The usage of filler metals with lower melting points makes the brazing process easier in practical application. The present work tries to develop Cu-Sn filler alloys containing Ti with low melting points, and clarify the mechanism of brazing SiC using the alloys.

2. Experimental Procedure

The brazing alloys used were two series of Cu-Xat% Sn-21at% Ti and Cu-16.5at% Sn-Xat% Ti. Pressureless sintered SiC contained a few percent alumina as sintering aid. SiC of 6mm diameter and 3mm thickness to SiC of 15mm diameter and 3mm thickness were brazed using the brazing alloys of 6mm diameter and 0.1mm thickness in vacuum. The melting points of the alloys were measured using a differential thermal analyzer. The brazing temperature and time were 1123 K to 1273 K and 1.8 ks in vacuum. The brazing temperature using the Cu-Sn-Ti alloys could be lowered to 1123 K though the conventional brazing temperature using Cu-Ag-Ti alloys was 1223 K. The microstructure and elemental analysis of SiC/SiC joints were investigated by using an electron probe microanalyser and X-ray diffractometer. The strength of SiC/SiC joints was evaluated by fracture shear testing with a cross head speed of $1.7x10^{-5}$m/s.

3. Results and Discussion

Fig. 1 shows the change in strength of a SiC joint with Ti content in the Cu-16.5at%Sn alloys at the brazing temperature of 1223 K for 1.8 ks. The strength of SiC joint brazed with Cu-16.5at%Sn alloys presents a maximum at 5.6 at%Ti. The microstructure of the SiC joint brazed with Cu-16.5at%Sn-5.6at%Ti alloys is

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increases the strength of the SiC joint. The microstructure of the SiC joint brazed with Cu-16.5at%Sn-5.6at%Ti alloys is shown in **Fig. 2**. The reaction zone containing Ti is formed at the interface between SiC and the joining layer. The quantitative analyses and X-ray diffraction indicated that the interface reaction zone is composed of a TiC layer zone adjacent to SiC and Ti_5Si_3 granules. The matrix of the joining layers are the eutectoid structures composed of α-Cu solid solution containing Si and ε-Cu_4Sn. The eutectoid structure of the matrix changes with the addition of 5.6 at% Ti or more. The fine structure of the eutectoid matrix in the joining layer accounts for the maximum strength of the SiC joint. **Fig. 3** shows the brazing time dependence of the strength of three SiC joints brazed with Cu-16.5at%Sn-3.7, 5.6 and 7.9 at%Ti alloys. The Cu-16.5at%Sn-5.6at% Ti alloy among the three alloys yields the highest values of the strength. Those SiC joints yield the maximum strength at the brazing time of 1.8 ks. The change in number and size of Ti_5Si_3 near the interface accounts for the change of strength with the brazing time. **Figs. 4 and 5** show the histograms of number and size of Ti_5Si_3 formed near the interface between SiC between SiC and the joining layer with the brazing time and the joining layer for the SiC joint brazed at 1223 K for 1.8 ks and 7.2 ks using Cu-16.5 at%Sn-5.6 at%Ti alloy. Although a number of fine Ti_5Si_3 are formed at the interface of the SiC joint brazed at 1.8 ks, the many bigger Ti5Si3 are formed at the joint brazed at 7.2 ks. The many brittle intermetallics near the interface cause to reduce the strength of the SiC joint.

For comparison, the Ti content dependence of strength brazed with Cu-Ti alloys at 1373 k for 1.8 ks is included in the figure 1. Although the strength of SiC joint yields a maximum at 5.6at%Ti, the Ti addition to Cu-Ti alloys continuously improves the strength of the SiC joint. **Fig. 6** represents the comparison of the microstructures between SiC joint brazed with Cu-5.2at%Ti alloy and the SiC joint brazed with Cu-16.5at%Sn-5.6at%Ti alloy. Cu in the Cu-5.2at%Ti alloy penetrates into the SiC. The

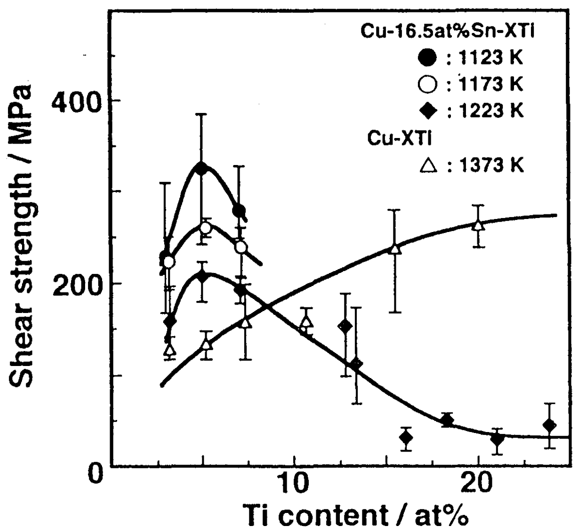

Fig. 1 Change in strength of SiC/SiC joint with Ti content at Cu-16.5at% Sn alloys.

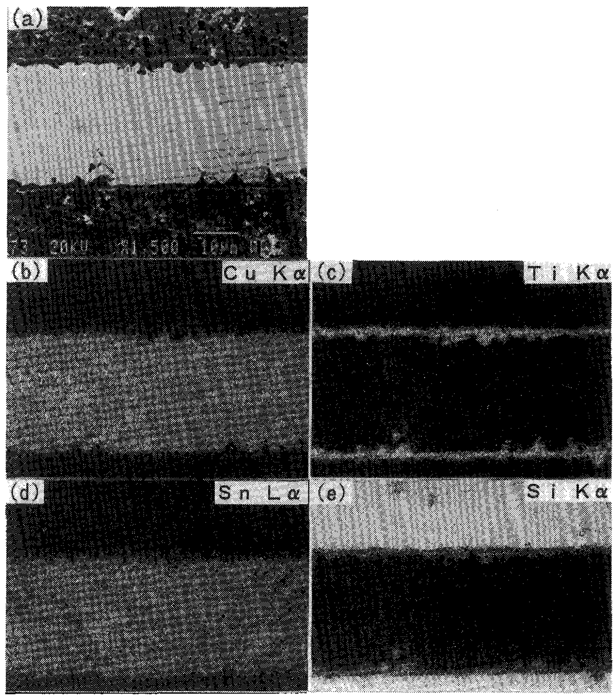

Fig. 2 Micostrucutre and X-ray image analyses of elements for SiC/SiC joint brazed with Cu-16.5 at%Sn-5.6at%Ti alloy.

copper reacts with SiC and forms the Cu silicides zones between the SiC and joining layer in **Fig. 6a**. On the other hand, Cu in the Cu-16.5 at% Sn-5.6 at%Ti does not penetrate into SiC as shown in **Fig. 6b**. The interface is composed of only the TiC zone and Ti_5Si_3 granules. In other words, Sn in the Cu-Sn has the role of preventing the reaction of Cu with SiC.

A decrease in brazing temperature improves the strength of the SiC joint as shown in Fig. 1. The change in the morphology of microstructure of the reaction zone leads to the increase in the joining strength of the joint. Ti_5Si_3 in a reaction zone shows the separated granules at the lower brazing temperature of 1123 K. The Ti_5Si_3 forms the layer zone at the higher brazing temperature of 1223 K. The layer zone of Ti_5Si_3 accounts for the lower strength of the SiC joint.

The SiC joints brazed with Cu-16.5 at% Sn containing of 20 at%Ti or more possess the lower strength of SiC joint in Fig. 1. Large amounts of intermetallics are formed by adding Ti content of 21at% is seen in **Fig. 7**.

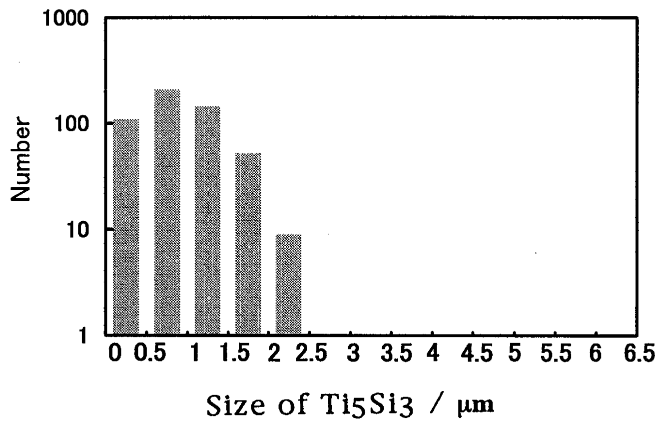

Fig. 4 Histogram of number and size for Ti_5Si_3 in SiC joint with Cu-15.5at%Sn-5.6at%Ti brazed at 1223 K for 1.8 ks.

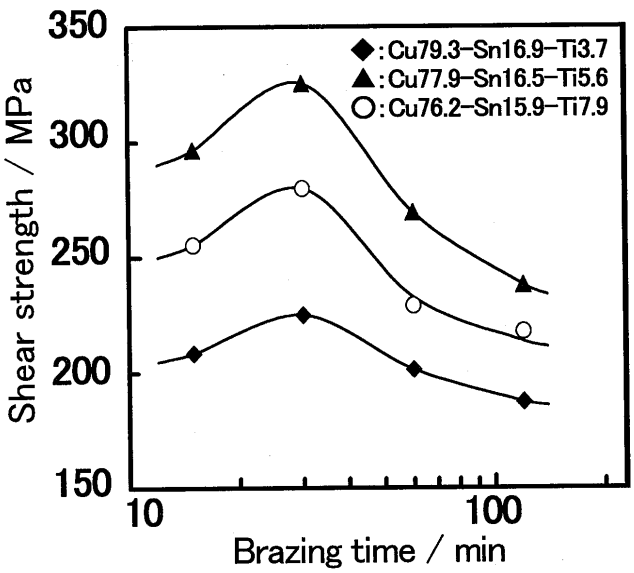

Fig. 3 Brazing time dependence of SiC joint with Cu-16.5at%Sn-3.7, 5.6, 7.9at%Ti alloys brazed at 1223 K.

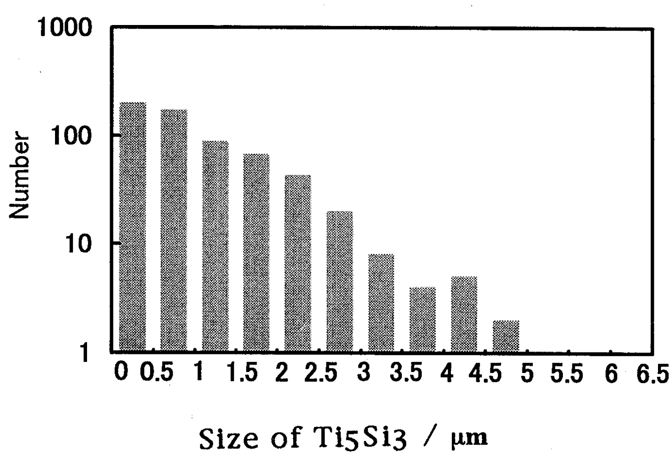

Fig. 5 Histogram of number and size for Ti_5Si_3 in SiC joint with Cu-16.5at%Sn-5.6at%Ti brazed at 1223 K for 7.2 ks.

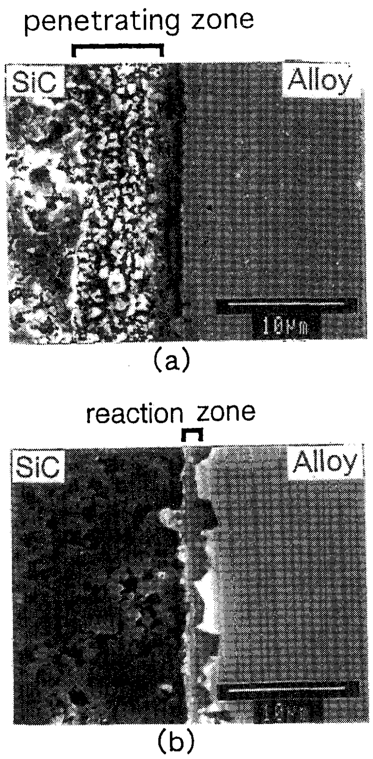

Fig. 6 Comparison of microstrucutres of X-ray image analyses of elements for SiC joint brazed with Cu-16.5at%Ti alloy.-21at%Ti alloy.

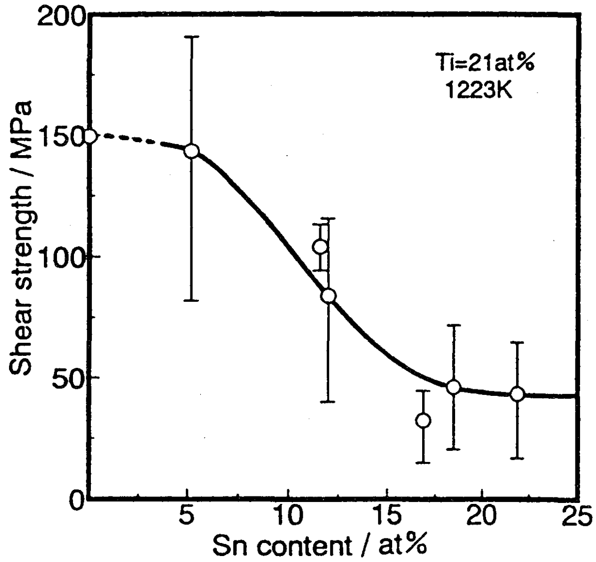

Fig. 8 Change in strength of SiC/SiC joint with Sn content at Cu-21at%Ti alloys..

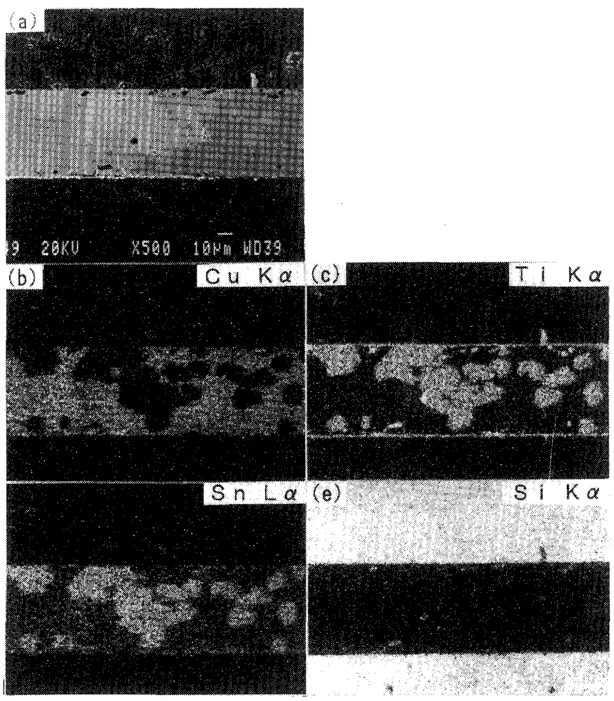

Fig. 7 Microstrucutre and X-ray image analyses of ellements for SiC/SiC joint brazed with Cu-16.5at%Sn-21at%Ti alloy.

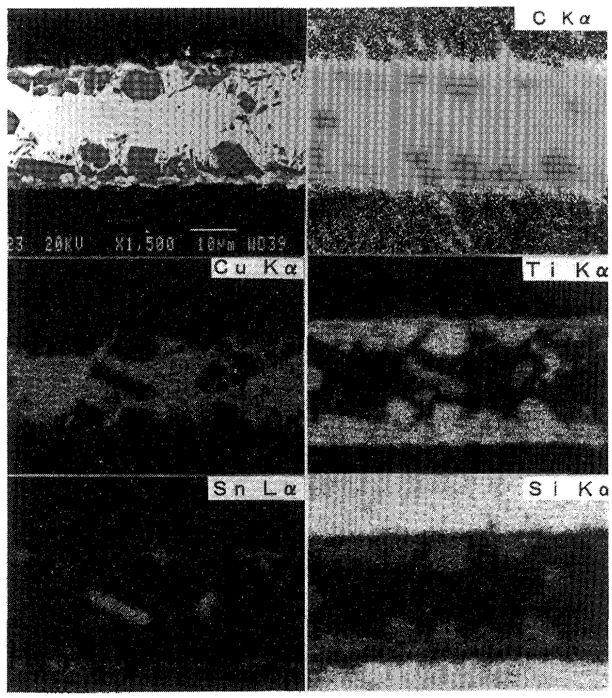

Fig. 9 Microstrucuture and X-ray image analyses of elements for SiC/SiC joint brazed with Cu-5.2at%Sn-21at%Ti alloy.

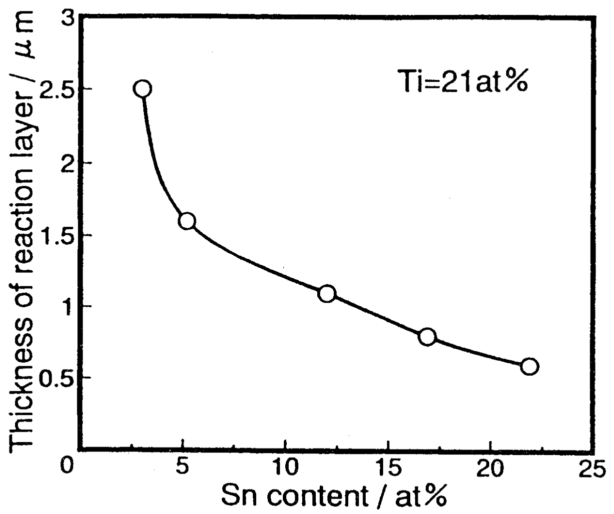

Fig. 10 Change in thickness of reaction zone of SiC/SiC joint with Sn content in Cu-21at%Ti alloys.

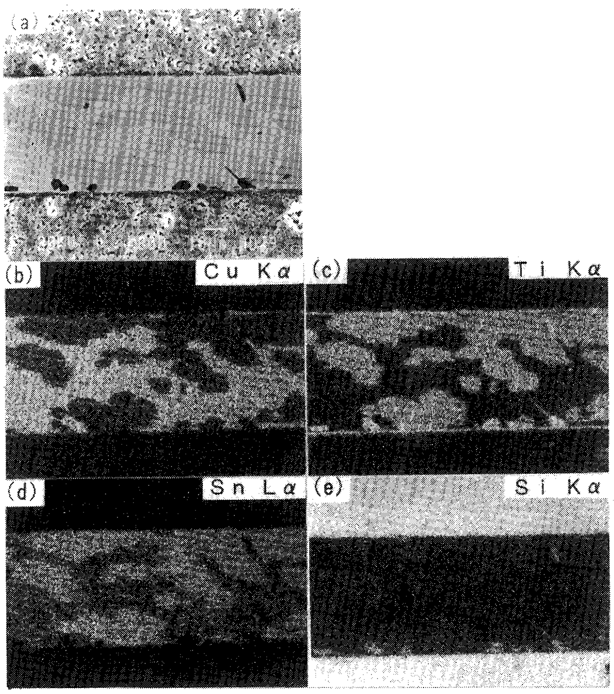

Fig. 11 Microstrucutre and X-ray image analyses of elements for SiC/SiC joint brazed with Cu-21.9at%Sn-21at%Ti alloy.

The reduction of strength of the SiC joint brazed with Cu-21at%Ti alloys is also seen by increasing Sn content in the alloys as shown in **Fig. 8**. The reaction zones are composed of TiC and Ti_5Si_3 at the interface between SiC and the joining layer in the SiC joint with Cu-5.2at%Sn-21at%Ti alloy in **Fig. 9**. **Fig. 10** shows that the addition of Sn to Cu-21at%Ti alloys definitely reduces the thickness of the reaction phase zone. The matrix is composed of many intermetallic granules containing Cu, Sn and Ti as shown in **Fig. 11**. The quantitative composition of the intermetallics indicates the chemical formula of CuSn3Ti5. The large amounts of the brittle intermetallics reduce the strength of the SiC joint. The Sn addition to Cu-5.6at%Ti alloys possess the two opposite roles. First, small amounts of Sn in Cu alloys prevents the direct reaction of Cu with SiC. The reaction zone of TiC and Ti_5Si_3 at the interface between SiC and the joining layer strongly connects the interface. Secondly, the excess addition of Sn to Cu-Ti alloys causes the formation of the brittle intermetallics of $CuSn_3Ti_5$ in the joining layer. The formation of the intermetallics reduce the amount of TiC and Ti_5Si_3 in the reaction zone near the interface between the SiC and the joining layer, and lowers the thickness of the reaction zone as shown in Fig. 10. The decrease in the reaction zone causes the reduction of the strength of the SiC joint as shown in Fig. 8. Furthermore, the formation of large amounts of $CuSn_3Ti_5$ intermetallics reduces the strength of the SiC joint.

4. Conclusions

Brazing of SiC to SiC was performed by using two series of Cu-Xat% Sn-21at% Ti and Cu-16.5at% Sn-Xat% Ti alloys at 1223 K for 1.8 ks in vacuum. The brazing mechanism was investigated by observing the interfacial microstructures, and measuring the fracture shear stress of the SiC joints.

At constant Sn content of 16.5 at%, the strength of SiC/SiC joint represents the maximum strength at the Ti content of 5.6at%Ti. The Sn in Cu-Ti alloys prevents the penetrating of molten copper into SiC, the reaction zone of TiC and Ti5Si3 formed at the interface between SiC and the joining layer and the eutectoid matrix of α-Cu and δ-Cu_4Sn accounts for the high strength of the SiC/SiC joint. In particular, many distribution of fine Ti_5Si_3 granules is attributable to the maximum strength of the joint. The excess addition of Sn and Ti to Cu forms the brittle intermetallic of CuSn3Ti5 in the joining layer and reduces the strength of the SiC joint.

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