Development of Cu-Sn or Cu-Zn Filler Metal for Joining Ceramics†

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Abstract

SiC was brazed to SiC using Cu-Sn-Ti filler metals with low melting points. At a constant Sn content of 16.5 at% the strength of the SiC/SiC joint brazed at 1123 K in vacuum shows a maximum at 5.6 at%Ti , and decreases with an increasing content of Ti in the alloys. The fine TiC and Ti_5Si_3 precipitates distribute densely at the SiC/the alloy interface. The excess amounts of Sn and Ti in at% the alloys form a brittle intermetallic of $CuSn_3Ti_5$ in the joining layer of SiC/SiC, and affect the strength of the joint. SiC was also brazed to SiC using Cu-30at%Zn-Ti at the low brazing temperature of 1223 K in vacuum.

KEY WORDS: (Brazing) (Ceramics) (SiC) (Si_3N_4) (Cu-Sn) (Cu-Zn) (Titanium)

1. Introduction

Silicon base ceramics are candidate materials for structural ceramics. Joinig of ceramics enlarges possible engineering applications [1]. It is known that copper base alloys containing Ti easily wet SiC or Si_3N_4, 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 [4-9] alloys. The usage of filler metals with lower melting points makes the brazing process easier in practical applications. The present work tries to develop Cu-Sn or Cu-Zn filler alloys containing Ti with low melting points, and clarifies the mechanism of brazing SiC or Si_3N_4 using those alloys.

2. Experimental Procedure

The liquidus surface and melting points of Cu-Sn-Ti ternary alloys were investigated by observing microstrucutre and measuring DSC curves of melted alloys. The brazing alloys used were Cu-16.5at%Sn-Xat%Ti. Pressureless sintered SiC contained a few percent of alumina as a sintering aid. SiC of 6mm diameter and 3 mm thickness to SiC of 15mm and 3 mm thickness were brazed using Cu-Sn-Ti filler metals of 0.1 mm thickness in vacuum at high temperture. Si_3N_4 was also brazed using Cu-Zn-Ti filler metals. The microstructure and elemental analysis of SiC/SiC or Si_3N_4/Si_3N_4 joints were investigated by using an electron probe microanalyser and X-ray diffractometer. The strength of SiC/SiC or Si_3N_4/Si_3N_4 joints was evaliuated by fracture shear testing with a cross head speed of 1.7x10-5m/s.

3. Results and Discussion

Fig. 1 shows the liquidus surface of the Cu-Sn-Ti ternary system. Two intermetallics of CuSnTi and $CuSn_3Ti_5$ are formed in the system, and the liquidus surface of $CuSn_3Ti_5$ is wide from the lower Ti content of 10 at% to the higher Ti content of 60 at%. At a constant Sn content of 15 at% the addition of Ti definitely decreases from 113 K for the binary Cu-15at%Sn alloy to 1046 K for 15 at%Ti. The further addition of Ti forms the intermetallic of $CuSn_3Ti_5$. At a constant Ti content of 20 at% the addition of Sn also reduces the melting points of alloys from 1203 K for binary Cu-20at%Ti alloy to 1116 K for 20at%Sn. At a constant Sn content of 16.5 at% the strength of the SiC/SiC joint brazed at 1123 K in vacuum shows a maximu at 5.6 at%Ti, and decreases with the further increase of Ti in the alloys. The fine TiC and Ti_5Si_3 precipitates distribute dense $CuSn_3Ti_5$ in the joining layer of SiC/SiC, and affect the strength of the joint.

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Fig. 2 shows the brazing time dependence of the strength of three SiC joint s brazed with Cu-16.5at%Sn-3.7, 5.6 and 7.9at%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 1.8ks. The change in number and size of TiC and Ti_5Si_3 precipitates near the interface affects the strength of SiC joint. The many fine TiC and Ti_5Si_3 precipitates near the interface at the shorter brazing timne accounts for the higher strength of SiC joint, and larger TiC and Ti5Si3 particles at the longer brazing time reduce the strength of the SiC joint.

A series of $(CuZn)_{(100-X)}Zn_{(X)}$ (X=0-20at%) fillers were prepared for brazing Si_3N_4 to Si_3N_4 in vacuum. The filler metals were prepared by conbining Cu-Zn foil 0.1 mm thick and Ti foil 20 mm thick. The content of Zn in Cu-Zn foil used was 35 mass% Zn, and the nominal content of Zn in the filler metals changed from 32 at% to 29 at% with increasing Ti content from 5 at%Ti to 20at%Ti. The lap of Si_3N_4 to Si_3N_4 pellets was brazed with the Cu-Zn-Ti filler metals in vacuum, where the sizes of Si_3N_4 were 6 mm diameter and 4 mm thick distribute densely at the SiC/the alloy interface.

The addition of Zn decreases the melting points of Cu-Zn-Ti alloys with Ti contents up to 20 at% and also the brazing temperature of the Si_3N_4[10]. The brazing temperature of 1223 K for Si_3N_4 joint with the Cu-Zn-Ti filler metals was comparably as low as the brazing temperature of Ag containing Cu-Ti-Ag filler metals[3,8,9], while the higher brazing temperature of Si_3N_4 joint with Cu-Ti was 1373 K[2]. The joining of the Si_3N_4 to Si_3N_4 was done at the brazing condition of 1223K for 1.8 ks.

Fig. 3 shows the change of microstructure of Si_3N_4/Si_3N_4 joint with a Ti content of $(CuZn)_{(100-X)}Ti_{(X)}$ in the filler metals. The amounts of reaction products in the joining layer of Si_3N_4 joints increase with the Ti content in the filler metals. The reaction zones are divided to the layer zone beside Si_3N_4 and the other zones containing granular precipitates in the central joining zone. The reaction zones in the Si_3N_4 joint brazed with $(CuZn)_{85}Ti_{15}$ filler metal were analyzed by electron probe analyser as shown in **Fig. 4**. The layer zone II beside Si_3N_4 (I) with N and Si with high Ti content was analysed as a mixture of TiN, Ti_2N and Ti_5Si_3 by X-ray diffraction analysis. Ti_2N was formed by the decomposition of TiN during cooling after brazing . The main matrix marked by IV and VI is Cu solid solution matrix marked by IV and VI is Cu solid solution containing Zn content of 18at%. The matrix in the central joining layer contains the precipitates of Ti_2N, TiN and Ti_5Si_3 (III and V). An increase in the Ti content of the filler metal causes an increase in the thickness of reaction zones comprised of TiN or Ti_2N and Ti_5Si_3 beside Si_3N_4, and also the amounts of precipitates of TiN or Ti_2N in the matrix in the joining layer.

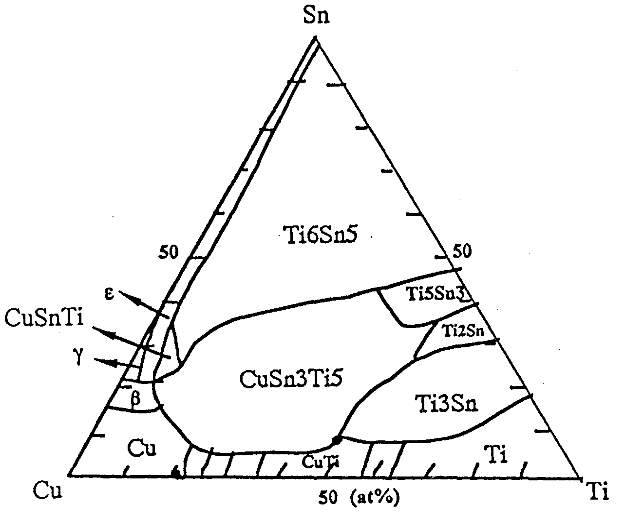

Fig. 1 Liquidus surface of the Cu-Sn-Ti ternary system.

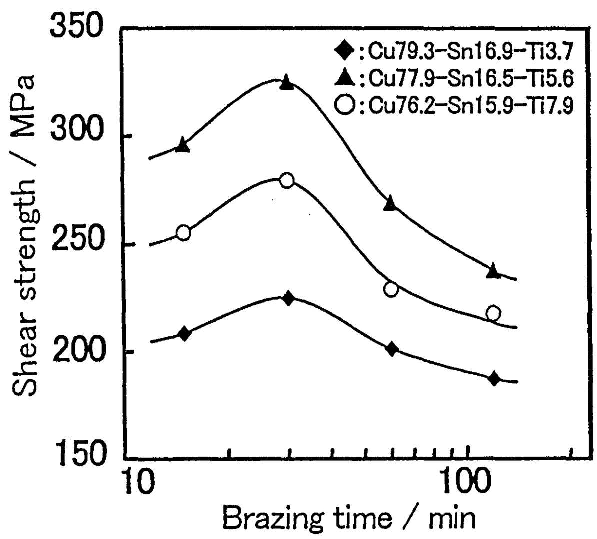

Fig. 2 Brazing temperature dependence of stremgth of SiC joints.

The content of Zn in the matrix of the Cu-Zn solid solution in the central joining layer became lower than the initial Zn content in the brazing filler metal. The zinc in the filler metal evaporated during brazing in vacuum. For instance, the Zn content in the joint brazed with $(CuZn)_{85}Ti_{15}$ became 14at%, though the initial Zn content was 29.2at%. The melting point of the Cu-Zn-Ti filler metal becomes lower with the increase in the Ti content in the filler metals as indicated in the Cu-Zn-Ti phase diagram [5].

The increase in Ti content increases the amounts of reaction phases in the joining layer and the increase in the Ti content up to 15at% increases the thickness of the reaction zones. An increase in Ti content of 15at% or more reduces the thickness of the reaction zone. The excess content of Ti in the joining layer increases the flowability of the filler, and the filler metal flows out from the joining layer. This flowing-out of the molten filler metal reduces the thickness of the reaction zones and the joining layer in the joint. These changes in amounts of reaction phases and the joining layer are correlated with the change in the strength of the joint as shown in **Fig. 5**. The increase in the reaction zones with the increase in Ti content up to 10 at% in the filler metals improves the strength of the Si3N4, and the decrease in the joining layer for the Ti content of 15 at% or more also definitely increases the strength of the joint as shown in Fig. 5.

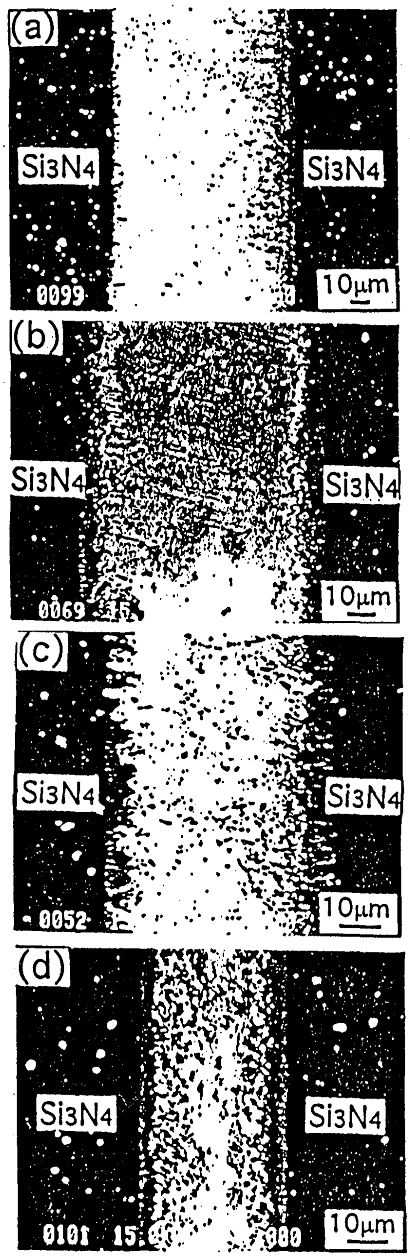

Fig. 3 Chamge in microstructure of Si_3N_4/Si_3N_4 joint with a Ti content of $(CuZn)_{100-X}-Ti_X$.

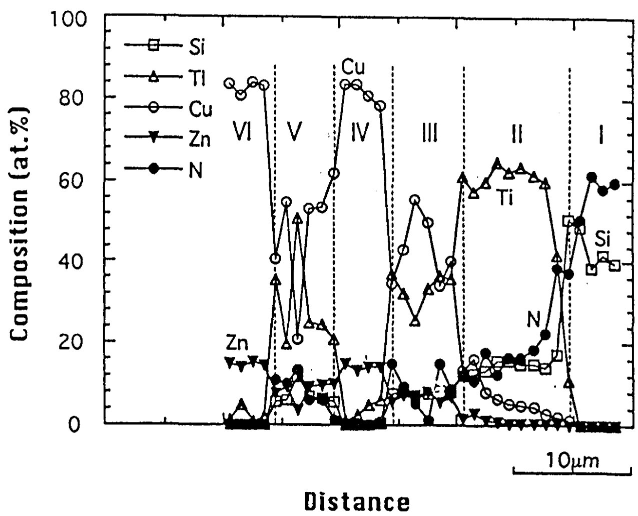

Fig. 4 Line analyses of elements in Si_3N_4 joint brazed with $(CuZn)_{85}Ti_{15}$ filler metals at 1223 K for 1.8ks.

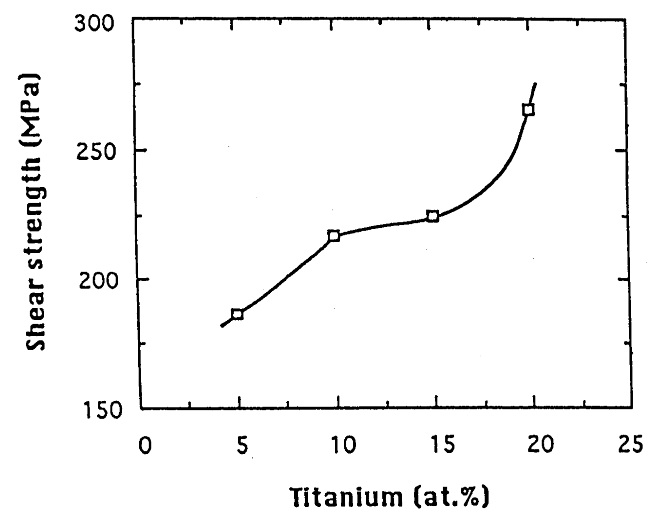

Fig. 5 Change in stremgth of Si_3N_4 joint with Ti content in (CuZn)Ti filler metals.

4. Conclusions

The filler metals of Cu-Sn and Cu-Zn containing Ti with low melting points were prepared for joining SiC or Si_3N_4. At a constant Sn content of 16.5 at% the strength of the SiC/SiC joint brazed at 1123 K in vacuum shows a maximum at 5.6 at%Ti , and decreases with a further content of Ti in the alloys. The fine TiC and Ti_5Si_3 precipitates distribute densely at the SiC/alloy interface. 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 joint, and reduce the strength of the joint. The addition of Zn to Cu-Ti filler metals also lowers the brazing temperature of Si_3N_4 joint to 1223 K. Ti in the Cu-Zn-Ti actively reacts with Si_3N_4, and forms a reaction layer zone and precipitates of TiN or Ti_2N and Ti_5Si_3 in the joining layer. These compounds results in the sound joining of Si_3N_4.

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