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Interfacial microstructure of Si_3N_4/Si_3N_4 brazing joint with Cu–Zn–Ti filler alloy

J. Zhang [a,*], X.M. Zhang [a], Y. Zhou [a], M. Naka [b], Atroshenko Svetlana [c]

[a] School of Materials Science and Engineering, Harbin Institute of Technology, Harbin 150001, China
[b] Joining and Welding Research Institute, Osaka University, Ibaraki, Osaka 567-0047, Japan
[c] Faculty of Mathematics and Mechanics, Saint-Petersburg State University, Russia

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Abstract

In this study, Si_3N_4 ceramic was jointed by a brazing technique with a Cu–Zn–Ti filler alloy. The interfacial microstructure between Si_3N_4 ceramic and filler alloy in the Si_3N_4/Si_3N_4 joint was observed and analyzed by using electron-probe microanalysis, X-ray diffraction and transmission electron microscopy. The results indicate that there are two reaction layers at the ceramic/filler interface in the joint, which was obtained by brazing at a temperature and holding time of 1223 K and 15 min, respectively. The layer nearby the Si_3N_4 ceramic is a TiN layer with an average grain size of 100 nm, and the layer nearby the filler alloy is a $Ti_5Si_3N_x$ layer with an average grain size of 1–2 μm. Thickness of the TiN and $Ti_5Si_3N_x$ layers is about 1 μm and 10 μm, respectively. The formation mechanism of the reaction layers was discussed. A model showing the microstructure from Si_3N_4 ceramic to filler alloy in the Si_3N_4/Si_3N_4 joint was provided as: Si_3N_4 ceramic/TiN reaction layer/$Ti_5Si_3N_x$ reaction layer/Cu–Zn solution.

Keywords: Si_3N_4 ceramic; Brazing; Cu–Zn–Ti filler alloy; Interfacial microstructure

1. Introduction

Si_3N_4 ceramic is a promising structural material because of its excellent physical and mechanical properties. Welding techniques have been widely used in ceramic structure [1–5] because ceramics are usually very brittle and have lower machinability and formability. Active brazing is one of the widely accepted techniques for ceramic welding [6]. In order to get a high performance ceramic joint by the brazing technique, it is very important to use a suitable filler alloy which has a good wettability to ceramic substrate. Ti alloys have been widely used as filler alloys for brazing Si_3N_4 ceramic because Ti reacts with Si_3N_4 at high temperatures forming a TiN reaction layer at the ceramic/filler interface, which improves the wetting between Ti alloy filler alloy and Si_3N_4 ceramic. Therefore, various binary or ternary Ti alloys have been investigated as filler alloys to obtain ceramic joints with required properties [7–9].

It is well known that, during cooling from brazing temperature to room temperature, a residual thermal stress forms in the ceramic nearby the ceramic/filler interface because of the big difference of coefficient of thermal expansion between ceramic and metal alloy, resulting in a decrease of the properties of the ceramic joint [10]. The higher the brazing temperature, the larger the residual stress. In this investigation, a Cu–Zn–Ti alloy was used as the filler alloy for brazing Si_3N_4 ceramic. The addition of Cu and Zn in the filler alloy can decrease melting temperature of the filler alloy, so the brazing temperature and in turn the residual stress can be decreased [11].

Properties of the welding joint depend greatly on the microstructure of the joint, therefore, it is necessary to study the interfacial microstructure of the joint [12,13]. In this study, morphology and microstructure of various phases and their interfaces were observed and analyzed by means of electron-probe microanalysis (EPMA), X-ray diffraction (XRD) and transmission electron microscopy (TEM).

2. Materials and experimental procedures

Cu65Zn35 (wt.%) alloy foil with a thickness of 0.1 mm and pure Ti foil with a thickness of 20 μm were used as filler materials to braze the Si_3N_4 ceramic. Commercial Si_3N_4 ceramic samples with a size of Ø 6 mm × 4 mm were ground to a surface

* Corresponding author. Tel.: +86 451 86414234; fax: +86 451 86413922.
E-mail address: hitzhangjie@hit.edu.cn (J. Zhang).

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finish of $R_a = 30\,\mu m$, and then were cleaned together with the metal foils in a ultrasonic bath. The cleaned metal foils were put between two pieces of Si_3N_4 ceramic and then heated at a heating rate of 25 K/min to 1223 K in a vacuum of (1.33–1.67) × 10^{-3} Pa. The brazed sample was hold at the brazing temperature for 15 min under a pressure of 2×10^{-3} Pa and then cooled in the furnace. By adjusting the amount of the two kinds of foils, a filler alloy with a composition of (Cu, Zn)85Ti15 (at.%) will be formed in the joint between Si_3N_4 ceramics during the heating process.

The samples for XRD analysis were obtained by cutting and grinding the brazed sample parallel to the joint plane. Elemental distribution across the joint was measured by EPMA. Samples for TEM observation were made by the focused ion beam technique, and microstructure of the Si_3N_4/filler/Si_3N_4 joint was observed and analyzed by means of TEM.

3. Results and discussion

Fig. 1 shows the XRD result of the joint brazed at 1223 K for 15 min. It indicates that besides Si_3N_4 substrate and Cu-based solution filler alloy, three new phases (TiN, Ti_5Si_3 and Cu_2TiZn) are found in the joint. Fig. 2 shows the morphology and elemental analysis results of the joint. Fig. 2(a) shows a clear reaction layer between Si_3N_4 substrate and filler alloy. Fig. 2(b) shows some new phases in the center part of the joint. Distribution of Cu, Ti, Zn, Si and N across the joint was measured along the broken line in Fig. 2(b), and the results were shown in Fig. 2(c). It was found that the reaction layer between Si_3N_4 substrate and filler alloy contains mainly Ti, N and Si, indicating that this layer is composed of Ti–N and Ti–Si compounds. When the XRD results are considered and Ti–N, Ti–Si–N and Cu–Zn–Ti phase diagrams [14–16], are referred, it is presumed that the reaction layer between Si_3N_4 substrate and filler alloy contains Ti–N and Ti–Si or Ti–Si[N] reaction phases as show in Fig. 2(b) and Table 1 (point A).

In order to identify the new phases in the center of the joint, composition of some special point (marked by B, C and D in Fig. 2(b)) was measured and the results are shown in Table 1. The background subtraction and corrections have been done to the EPMA results shown in Table 1 for atomic number, absorption and fluorescence. The result of the point B indicates that Cu–Zn

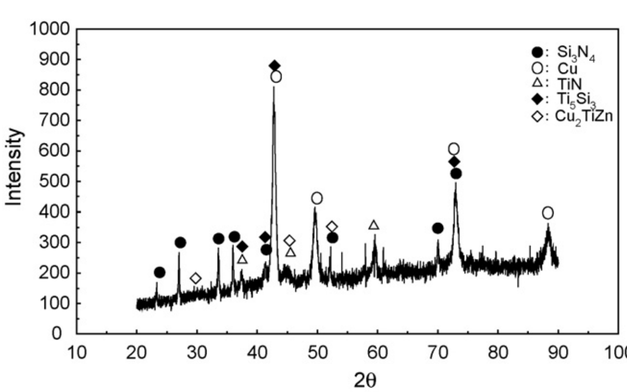

Fig. 1. XRD result of the Si_3N_4/Si_3N_4 joint brazed at 1223 K for 15 min.

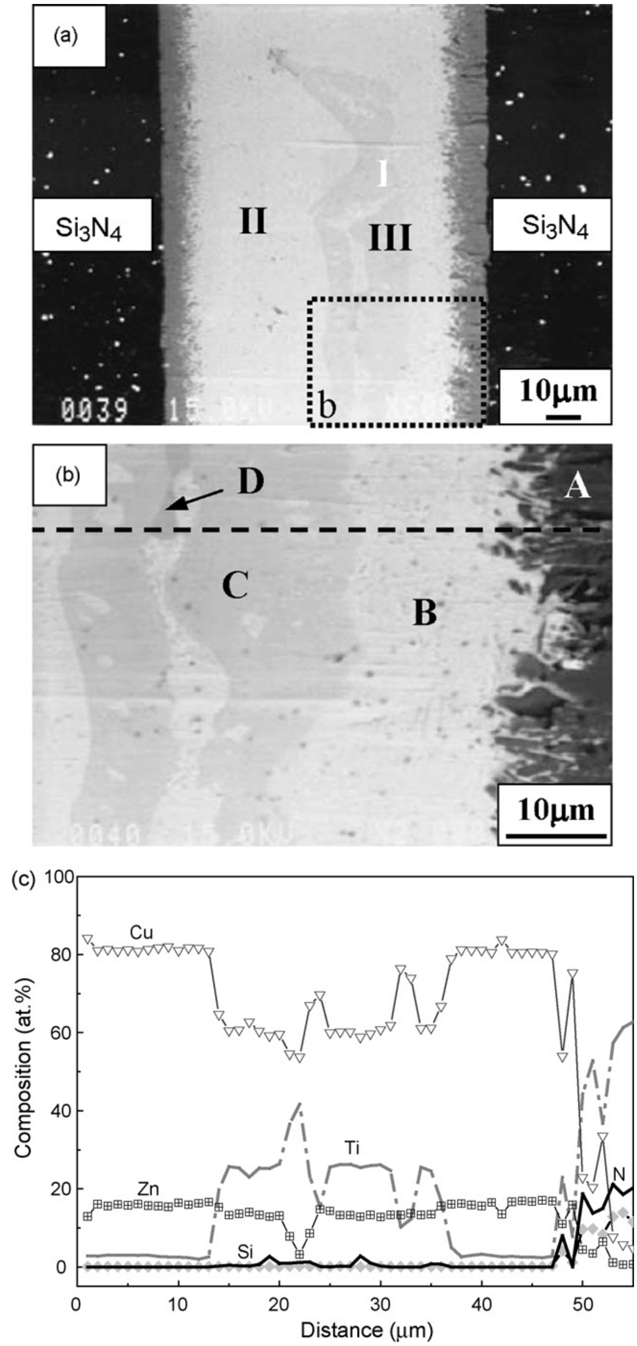

Fig. 2. Morphology of the joint and elemental distribution of Cu, Ti, Zn, Si and N across the joint along the broken line in (b): (a) morphology of a whole joint, (b) morphology of the joint by a higher magnification, and (c) elemental distribution.

Table 1
Composition of the points A, B, C and D shown in Fig. 2(b) (at.%)

Point	Si	Ti	Cu	Zn	N
A	7.688	66.625	6.671	0.832	18.156
B	0	3.125	80.510	16.365	0
C	0	26.197	60.785	13.019	0
D	0	43.829	52.922	3.019	0.445