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Interfacial Microstructure and Reaction Phases in Si_3N_4/Si_3N_4 Joints Brazed with Cu-Zn-Ti Filler Alloy

Jie Zhang*, Naka Masaaki **, Qingchang Meng* and Yu. Zhou***

Abstract

Si_3N_4 ceramic was jointed to itself using (CuZn)85Ti15 filler alloy at 1123-1323 K for 0.9 ks. TEM observation showed that a reaction zone of TiN and/or Ti_2N exists at the interface between the ceramic and filler alloy. When the brazing temperature is lower than 1223 K, the interfacial reaction zone is mainly composed of Ti_2N, which has cylindrical shape and is randomly orientate in the zone. There is a crystal orientation relationship between the Ti_2N in the reaction zone and the Cu in the Cu-Zn solid solution, which is: $\{110\}_{Ti2N}//\{420\}_{Cu}$, $<001>_{Ti2N}//<001>_{Cu}$. When the brazing temperature is higher than 1223 K, the interfacial zone is composed of TiN, which has plate shapes crossing each other.

KEY WORDS: (Si_3N_4 ceramic) (Cu-Zn-Ti filler alloy) (Brazing) (Microstructure)

1. Introduction

In recently 20 years, many studies have been focused on the techniques of ceramic joining because it can be used, not only for low-cost and high-reliability manufacturing of ceramic parts with complicated shapes, but also to repair ceramic parts with cracks. Active metal brazing is widely investigated because it is a simple process to obtain high strength ceramic joints with different shapes and sizes.

Most of the current researches about the active brazing of Si_3N_4 ceramic have focused on the following three points[1-3]: (1) the effect of brazing parameters on the microstructure and properties of the joints; (2) the analysis of residual stress at the joint interface; (3) the behavior of the interfacial reaction and the examination of the reaction products. This work studies microstructure and reaction phases in the Si_3N_4/Si_3N_4 joint with Cu-Zn-Ti filler alloy to observe and analyze the relationship between the different phases at the joint.

2. Experimental Procedures

Cu-Zn-Ti containing 15 at. % Ti was used as filler alloy to braze the Si_3N_4 to Si_3N_4 ceramic. The brazing process was carried out in a vacuum of $1.33\text{-}1.67\times 10^{-3}$ Pa for 0.9 ks under a pressure of 2×10^{-3} MPa. The brazing temperatures were from 1123 K to 1323 K with an interval of 50 K. The microstructure of the interface between Si_3N_4 ceramic and filler alloy was observed by TEM.

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3. Results

Fig.1 (a) is the TEM image showing the morphology of titanium nitride in the reaction zone between Si_3N_4 and the filler alloy obtained during brazing at 1173 K for 0.9 ks. Analysis results of the diffraction patterns (Fig.1 (b)) indicate that the titanium nitride in the reaction zone obtained at this temperature is Ti_2N. Fig. 2(a) also shows the morphology of the Ti_2N in the reaction zone obtained at the same brazing condition. From Fig.1 (a) and 2(a) it can be seen that the Ti_2N has a cylindrical shape with the length and diameter of 0.4-0.8 μm and 0.1-0.2 μm, respectively. Fig. 1(a) shows the morphology of the Ti_2N in the longitudinal direction, and Fig. 2(a) shows the cross section of the Ti_2N. Fig.2 (b) shows the electron energy loss spectrum (EELS) result of the Ti_2N shown in Fig. 2(a), indicating the existence of Ti and N in the phase in reaction zone.

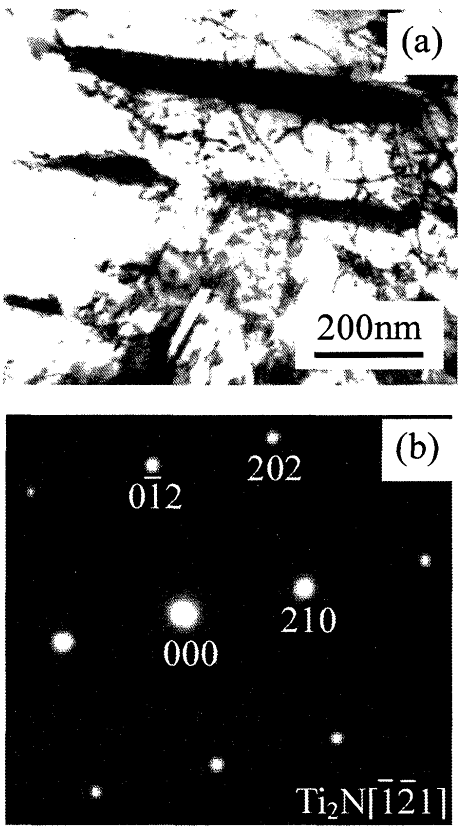

Fig. 1 TEM image showing the morphology of Ti_2N in the reaction zone obtained during brazing at 1173 K for 0.9 ks: (a) morphology of the Ti_2N, (b) diffraction patterns of the Ti_2N.

Fig.1 (a) and 2(a) also show that there is a high density of dislocations around the Ti_2N in the Cu-Zn solid solution. Because of the great difference of the coefficient of thermal expansion (CTE) between Ti_2N and Cu-Zn solid solution, interfacial stress arises during cooling from the brazing temperature, resulting in not only internal residual stress at the interface between Ti_2N and Cu-Zn solution but also local plastic deformation in the Cu-Zn solution and the formation of dislocations around the Ti_2N phases as shown in Fig.1 (a) and 2(a).

Fig.2(c) shows the diffraction patterns, which contain the patterns of both Cu-Zn solid solution in [001] direction and Ti_2N in [001] direction. Calculating and analyzing the results indicates that the large and small patterns correspond to Cu-Zn solid solution and Ti_2N respectively, and the indexing results are shown in Fig.2 (d), which clearly shows a crystal orientation relationship between the Cu matrix and the Ti_2N phase. The orientation relationship is $\{110\}_{Ti2N} // \{420\}_{Cu}$, $<001>_{Ti2N} // <001>_{Cu}$.

Fig.3 is the TEM image showing the morphology of titanium nitride in the reaction zone obtained during brazing at 1223 K for 0.9 ks. Compared with the Ti_2N shown in Fig.1 (a), the titanium nitride formed at 1223 K is much larger than Ti_2N. The titanium nitride shown in Fig.3 (a) has a plate shape with a thickness of 0.5-1.0μm, and they cross each other and are orientated randomly in the Cu-Zn solid solution. The diffraction patterns of the titanium nitride shown in Fig.3 (b) indicate that the titanium nitride formed at 1223 K is TiN. Because the Cu-Zn solid solution has been removed during the preparation of the TEM sample, it is not confirmed if there is crystal orientation relationship between TiN and Cu-Zn solid solution.

4. Discussion

During the brazing process, with increasing temperature, filler alloy gradually becomes molten and the Ti in the melt will diffuse to the interface between Si_3N_4 and filler alloy and concentrate near the surface of the Si_3N_4 ceramic, and then following reaction will take place to form titanium nitride [4]:

$$Si_3N_4(s) + 8Ti(l) = 4Ti_2N(s) + 3Si(s) \quad (1)$$

$$Si_3N_4(s) + 4Ti(l) = 4TiN(s) + 3Si(s) \quad (2)$$

According to the Ti-N binary alloy phase diagram [5], the Ti_2N phase is stable until 1373 K when the N content is 32.4-34at. %. With increasing temperature, the TiN phase becomes stable. The transformation temperature between Ti_2N and TiN decreases with

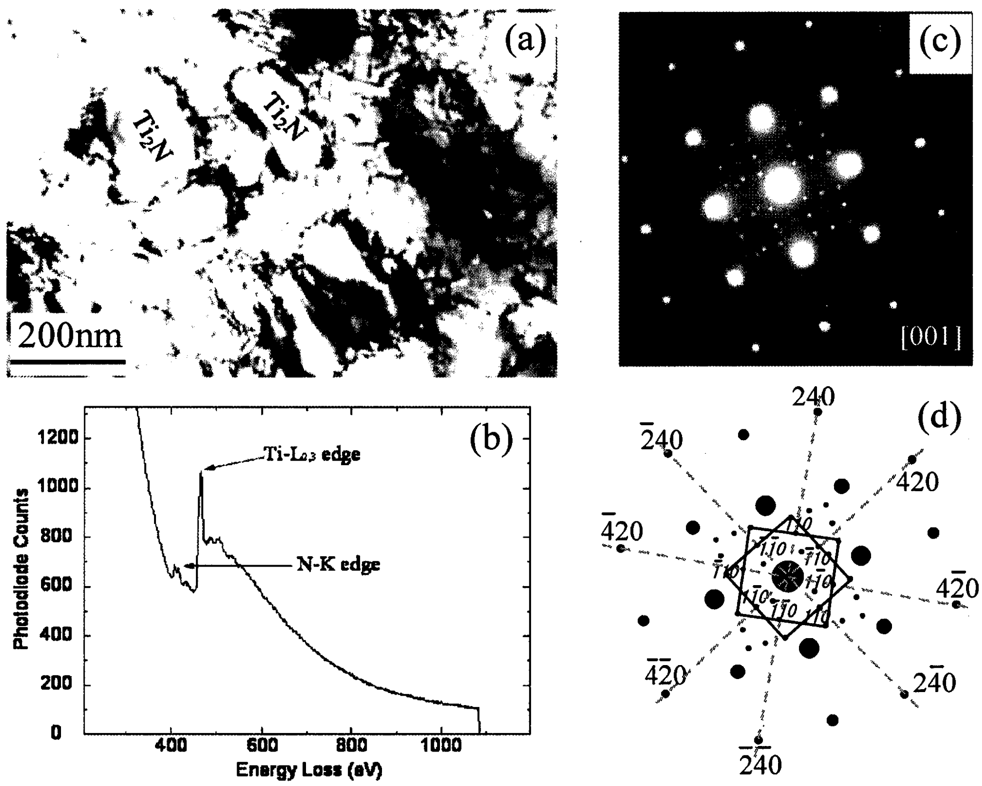

Fig. 2 TEM results showing the morphology and composition of the Ti_2N phase and the crystal orientation relationship between Ti_2N and Cu-Zn solid solution: (a) morphology of the cross section of the Ti_2N phase, (b) EELS results showing the existence of Ti and N in the Ti_2N phase, (c) diffraction patterns of both Ti_2N and Cu matrix, (d) indexing of the patterns.

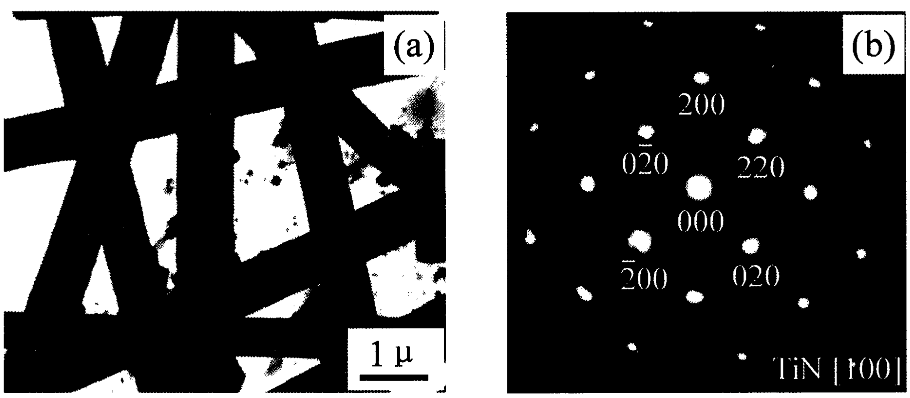

Fig. 3 TEM image showing the morphology of TiN in the reaction zone obtained during brazing at 1233 K for 0.9 ks: (a) morphology of the TiN, (b) diffraction patterns of the TiN.

increasing N content. In the brazing temperature range used in this investigation (1123-1323 K), Ti_2N phase will be formed according to reaction (1) at the interface between Si_3N_4 and filler alloy when the brazing temperature is lower. When the temperature surpasses 1223 K, TiN phase will be formed at the interface instead of Ti_2N phase.

5. Conclusions

A reaction zone of TiN and/or Ti_2N exists at the interface between the ceramic and filler alloy. When the brazing temperature is lower than 1223 K, the interfacial reaction zone is mainly composed of Ti_2N, which has a cylindrical shape and orientates randomly in the zone. There is a crystal orientation relationship between the Ti_2N in the reaction zone and the Cu in the Cu-Zn solid solution, which is: $\{110\}_{Ti2N}//\{420\}_{Cu}$, $<001>_{Ti2N}//<001>_{Cu}$. When the brazing temperature is higher than 1223 K, the interfacial zone is composed of TiN, which has plate shapes crossing each other.

Reference

[1] A. A. Prokopenko and V. S. Zhuravlrv, Journal of Materials Science Letters, pp. 2121-2123, V17 (1998).

[2] T. P. Swiler and R. E. Loehman, Acta Materials, pp. 4419-4424, V48 (2000).

[3] L. Esposito, A. Bellost and S. Landi, Journal of American ceramic society, pp. 3597-3604, V82 (1999).

[4] T. Shimoo, K. Okamura and S.Adachi, Journal of Materials Science, pp. 3031-3036, V32 (1997).

[5] Thaddeus B. Massalski, Binary alloy phase diagrams, American Society for Metal, Metals Park, Ohio 44073, pp. 970, 1986.